PHOTOIONIZATION OF ATOM BY INTENSE LASER FIELDS

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ABSTRACT

Photoionization of Atom by Intense Laser Fields

This research is devoted to a detailed study of the interaction between atom and intense laser field, which mainly focusing on the photoionization spectra of hydrogenlike atom. The main theoretical model which is involved in order to describe the photoionization phenomenon is Keldysh's model. Firstly, the Keldysh's formalism is derived in details and its physical significance is explored. We have presented the extension of the Keldysh's theory further into a semianalytical expression to study the characteristics of ionization rate of atom by intense lasers. In particular, the generalization of Keldysh's model is obtained for linear polarized light from small photoelectron momentum to arbitrary value of momentum. By applying different type of the laser field, i.e. linear, circular and elliptical, we have shown the variation of spectrum of the ionization rate and compare the features of the exact rates with Keldysh analytical result as functions of frequency and electric field strength. Next, the Keldysh's model is further extended in order to describe the photoionization of hydrogenlike atom from arbitrary initial energy level, which is not only restricted to the initial ground state energy level. A general analytical expression for arbitrary $n00$ energy level is obtained where n is the principal quantum number. Meanwhile, semianalytical expression is obtained for arbitrary nlm energy level where l is the azimuthal quantum number and m is the magnetic quantum number. Furthermore, we have also extended the Keldysh's theory to study the interaction between the hydrogenlike atoms with ultra intense laser where the relativistic effect is taken consideration in the model. The extension of the theoretical model into relativistic regime will provide a significant insight into the real world study since experimental works are deal with highly intense pulsed laser such as Ti:Sa laser for research study. The theory developed in this research will particularly benefit the future development of attoseconds laser, thus providing new tools for imaging and requiring further the development of electron control through intense light-matter interaction.

ABSTRAK

Pengionan Atom secara Foto dalam Medan Laser Sengit

Kajian ini ditumpukan kepada analisis interaksi antara atom dan medan laser sengit secara terperinci, di mana fokus utama diberikan kepada spektra pengionan hidrogenik atom secara foto. Model teori utama yang terlibat untuk menjelaskan fenomena pengionan secara foto ini adalah model Keldysh. Pertamanya, formalisme Keldysh diterbitkan secara terperinci dan kepentingan fizikalnya diterokai. Kami telah mempersembahkan perlanjutan model Keldysh ini kepada unggapan analisis separa untuk mengkaji ciri-ciri kadar pengionan atom dalam medan laser sengit. Khususnya, model Keldysh telah digeneralisasi untuk cahaya polarisasi linear daripada momentum foto elektron kecil kepada sebarang nilai momentum. Dengan mengaplikasikan medan laser yang berlainan, misalnya linear, bulatan dan elips, kami telah menunjukkan perubahan spektra daripada kadar pengionan dan membuat perbandingan antara kadar tepat dengan unggapan analisasi Keldysh secara fungsi frekuensi dan kekuatan medan elektrik. Seterusnya, model Keldysh telah dilanjutkan sekali lagi untuk menjelaskan pengionan hidrogenik atom secara foto daripada sebarang tahap tenaga awal, di mana tidak tertakluk kepada tahap tenaga dasar sahaja. Unggapan analisasi umum untuk sebarang tahap tenaga $n00$ telah didapati di mana n adalah nombor kuantum utama. Sementara itu, unggapan analisasi separa telah didapati untuk sebarang tahap tenaga nlm di mana l adalah nombor kuantum azimuthal dan m adalah nombor kuantum magnetik. Tambahan pula, kami juga melanjutkan teori Keldysh untuk mengkaji interaksi antara hidrogenik atom dengan laser berkuasa sengit di mana faktor kerelatifan diambil kira dalam pertimbangan. Perlanjutan model teori ini kepada bidang kerelatifan menyediakan pengetahuan fizikal yang penting kepada kajian dunia sebenar di mana kajian eksperimen biasanya berurusan dengan laser gelombang nadi untuk kajian laser sengit. Teori yang dibangunkan daripada kajian ini akan memberi manfaat kepada pembangunan masa depan kepada laser attoseconds, justeru menyediakan peralatan baru untuk pengimejan dan memerlukan pembangunan pengawalan elektron daripada interaksi jasad-cahaya sengit.

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Namo Tassa Bhagavato Arahato Samma-Sambuddhassa.

Veneration to the Blessed One, the Worthy One, the supreme perfect Awakened One.

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CHAPTER 1

INTRODUCTION

1.1 Introduction to Strong Field Ionization

Over the past few decades, the invention of laser has brought a remarkable advancement (Raymond Ooi & Seow, 2013) for the development of technologies such as femtosecond mid-infrared lasers (Popmintchev et al., 2012), intense few-cycle pulse (Hu & Collins, 2005), carrier-to-envelope phase control (Jones et al., 2000) and extreme ultraviolet laser (Richter et al., 2009). The recent technologies dates back the past 15 years such as X-ray free electron laser (Yu et al., 2000), attosecond pulses (Antoine, L'Huillier, & Lewenstein, 1996), high harmonic generation (Bandrauk & Shon, 2002) and molecular imaging (Peters et al., 2011) adopt the non-perturbative theory as the main theoretical engine. Strong field ionization of atom can no longer be described by the ordinary perturbation theory, loose definition since the ionization process requires multiphotons to be absorbed or emitted. In the past decade, the research by H. R. Reiss (Reiss, 1992) had introduce a new range of parameter which is the ratio of ponderomotive energy to the photon energy. This familiar metrics is defined as $z \equiv U_p/\hbar\omega$ where the ponderomotive energy

$$U_p = E^2/4\omega^2 \tag{1.1}$$

in atomic unit is the cycle-average kinetic energy of a free electron in an intense laser field with the laser amplitude E and the laser frequency ω. The z-value is having the dimension which is proportional to ω^{-3} and the physical meaning of this particular value gives the idea of how much is the photons exchanged during the interaction between the atom and the intense laser field. The invention of the intense femtosecond mid-infrared laser enable the finding of the z-value of several hundreds. Later on, following by the invention of X-ray free electron laser (XFEL) (Huang & Kim, 2007) allows the sequential removal of all the electrons from the parent atom although the particular z-value is practically zero.

In this field of research, strong field ionization requires a certain amount of painstaking works where the experiments are driving all along the time while the theory is being developed. For instance in the earlier theoretical work, even the Schrodinger equation for eliminating the single electron from the binding force of the strong external electromagnetic field and also the Coulomb potential of the particular atom cannot be solved exactly. In 1987, a new effect which is known as Stark-induced Rydberg resonances was observed and reported by Freeman (Freeman et al., 1987). Later on, the theoretical treatment in order to describe this phenomenon had been established, which is known as the Floquet's theory (Rottke et al., 1994). However, the method is still depending on the perturbation theory where low intensity of the laser field and short wavelength are taken into consideration. Perturbation theory starts to lose its significance when as soon as the photon energy is smaller compared to the larger Stark shifts effect. On the contrary, the solution of Schrodinger equation has to seek for other approximations beyond perturbation theory such as Keldysh's theory (Keldysh, 1965), and later by Faisal's theory (Faisal, 1973) and also Reiss's theory (Reiss, 1980, 1992). Additionally, the detailed comparison between the three different approaches always known as the Keldysh-Faisal-Reiss (KFR) theory (Popov, 2004). By using the non-perturbative approach in Keldysh's theory, where high intensity of the laser field and long wavelength limit are taken into consideration, hence the total ionization (R. Ooi, Ho, & Bandrauk, 2012) (In the later chapter, it is denoted as w) is behaving like the rate of dc-tunneling.

The Keldysh theory of describing the simple tunneling ionization has gain its popularity over years; even so, in the recent review of Reiss mentions that the concept of the tunneling ionization has a limited range of applications. Tunneling ionization has been playing the main role in the strong field ionization until the new theory, "The simple man model" developed by Corkum in 1993 followed by the high harmonic generation theory by Lewenstine (Lewenstein, Balcou, Ivanov, L'Huillier, & Corkum, 1994) and Becker (Becker, Lohr, Kleber, & Lewenstein, 1997) lead the strong field ionization to a new era of laser, which is known as attosecond physics. Corkum's model provides a classical description of the electron in the strong electromagnetic field, tunnel to the continuum and recollide with the parent ion, hence resulting in the maximum energy,

$$N_m \hbar\omega = I_p + 3.17 U_p \tag{1.2}$$

where I_p is the ionization potential energy and U_p is the ponderomotive potential as defined above. Furthermore, in the later work of M. Lein incorporates recolliding electrons for molecular imaging (Lein, 2007; N. Milosevic, Corkum, & Brabec, 2004). These works provide important foundations leading to the development of the intense light matter interactions (Bandrauk & Yu, 1999; E. Leorin & Bandrauk, 2007).

More experimental works right after the establishment of the theoretical models lead to the generation of high-energy attosecond light sources (G. Sansone & Nisoli, 2011) from gas. The attosecond light generation is based on the theory of high-harmonics generation where the electron is driven to the Volkov continuum state, returns and recollide to the parent ion (Lein & Rost, 2003), emitting a plateau of harmonics where the cutoff is located at $I_p + 3.17U_p$. A vast number of works are still trying to figure out the nature of the cutoff and whether other values beside $I_p + 3.17U_p$ are possible. Recent work by D. B. Milošević and A. F. Starace (B. & Starace, 1999) showed that the linearly polarized laser with the static field perpendicular to the laser field can induce a plateau towards high energy X-ray photons. K. J. Yuan and A. D. Bandrauk (K.-J. Yuan & Bandrauk, 2010, 2011b) had performed numerical results that showed the molecular high-order harmonic generation (Kamta & Bandrauk, 2005) can have the maximum elliptically polarized harmonic energies of $I_p + 13.5U_p$ (K.-J. Yuan & Bandrauk, 2010, 2011a), for certain internuclear distances and also relative pulse carrier envelope phase. The model shows that high-harmonic generation is not only contributed by the recollision of the electron to the parent ion of H_2^+ but also recollision with the neighbouring ion as well. Some work by D. B. Milošević, W. Becker, and R. Kopold has shown that circularly polarized harmonics (B., Becker, & Kopold, 2000) can be generated if the superposition of a linearly polarized laser field is orientated at an appropriate angle with a static electric field.

Since the past two decades, more applications (R. Ooi & Khoo, 2012; R. Ooi & Lee, 2013b; Ng & Ooi, 2013) in the strong field ionization have been developed base on the experiment findings such as Above Threshold Ionization (ATI), Multiphoton Ionization (MPI), resonant Multiphoton Ionization (RMPI) and the non-sequential double ionization. As a result of this, the new technology for instance, laser generating ultrafast light pulses becomes a surprise and a big gift by entering the millennium. Since then, more new tools are invented for the purpose of strong field investigation such as intense mid-infrared lasers, few-cycle pulses, carrier-to-envelope phase control and COLTRIMS devices. After the millennium year, following by a plethora of discovery such as attosecond generation and attochirp (Kohler, Keitel, & Hatsagortsyan, 2011), ion recoil momentum distributions (Weber et al., 2000), absolute phase effects (Stockman & Hewageegana, 2007), long-wavelength scaling of ATI (Corkum, Burnett, & Brunel, 1989), nonlinear optics (R. Ooi & Lee, 2013a) in the XUV (Van Dao, Teichmann, Davis, & Hannaford, 2008) and X-ray regime (Zhuang, Miranda, Kim, & Shen, 1999), non-sequential multiple ionization (Guo, Li, & Gibson, 1999), attosecond measurements of the ionization time delay and a universal strong field low-energy structure. In the evolution of strong field ionization, more and more concerns are focusing on controlling the electron dynamics so that new imaging tools can be developed base on high harmonics generation.

The new millennium imaging (Chen et al., 2009) tools are very important to explore the wonder of nature for our better understanding, for instance the chemical process. Since the chemical interaction is involving basically the electromagnetism force which is the transferring process of electron between atoms or molecules. In order to capture such a fast movement of chemical process (in the range around femtosecond, $\approx 10^{-15}s$), we need a faster imaging tool to breakthrough such as the attosecond generation ($\approx 10^{-18}s$). However, the invention of such a great technology requires a combination of knowledge from various aspects such as photoionization (R. Ooi et al., 2012), recombination (Zimmermann, Lein, & Rost, 2005) and re-collision (D. B. Milosevic & Ehlotzky, 2003) with parent ion in order to control the electron dynamics well. Hence, recent works start to focus back on these aspects such as photoionization and high harmonic generation (Clatterbuck et al., 2004) of different kind of laser fields, i.e. linear, circular and elliptical polarizations, photoelectron momentum distributions (K.-J. Yuan, Chelkowski, & Bandrauk, 2013); in order to provide a complete, well established theoretical model and also experimental justifications for a better understanding of the fundamental. By combining all these great works, the invention of this new millennium imaging (Teichmann, Chen, Dilanian, Hannaford, & Van Dao, 2010) tool can be achieved soon.

1.2 Motivations of the Research

As what we have discussed in the introduction, it is clear that the evolution of the strong field ionization and the world starts to concern on understanding the electron dynamics of an atom during the interaction with intense laser so that the controlling of electron can be done. However, many new findings recently are reporting the incompleteness of the past theories such as Keldysh and Ammosov-Keldysh-Delone (ADK) model. For example, in the recent work of Y. Z. Fu (Fu Yan-Zhuo, 2012), they found that the incompleteness of the ADK theory which overestimates the ionization rate and fails to give the correct ionization probability in the over-the-barrier (OBI regime). Without the establishment of a complete theoretical model, we can never fully understand physics phenomenon behind; as a result, a lot of new experiment findings meet bottleneck because these phenomenon cannot be well explained by the past theory. Consequently, we are motivated to develop a more general and well established theory for the better understanding of photoionization spectra of the hydrogen atom.

In our research, we study the Keldysh model in details and found that the incompleteness of the theory due to certain aspects, for example in the process of the interaction between hydrogen and the laser field, the laser field strength is not intense enough base on the new era perspective (perhaps it is considered very intense in the olden day). Besides, during the calculation, we found that Keldysh's assumption on the photoelectron is having a small momentum, where the low frequency of the laser field is considered since the direct proportionality of the photoelectron momentum and the laser frequency. In spite of that, Keldysh model has a limitation of setting the hydrogen ground state as the initial state of the tunneling ionization. Nevertheless, this is consider as an ideal case because the electron of the hydrogen atom cannot be in the exactly ground state in the real world. It might get excited to certain level due to some external factors with surroundings or interaction between other atoms.

In the later part, we will make an outline in details for each chapter and explain our method to overcome the issues as mentioned. This thesis is concerning on establish a more general and better theoretical model to describe the photoionization of atom in intense laser field for the sake of future technology such as the attosecond imaging tool.

1.3 Objectives

In this thesis, there are several important objectives to achieve. Firstly, we will study and discuss the past theoretical model of Keldysh on photoionization in details. Next, we set our focus to establish a general model on photoionization by considering arbitrary momentum of the photoelectron. Emphasize is made on the laser frequency and the momentum of photoelectron, comparison with our exact model and Keldysh model will be made and discussed in further. The next objective is to calculate and further extend the Keldysh's formalism to incorporate arbitrary energy level of the hydrogen atom to be the initial state of the photoionization. Last but not least, we will set the focus to consider the interaction of hydrogen atom with highly intense laser, where to relativistic effect is taken consideration in the system.

1.4 Thesis Organization and Outline

This thesis is roughly arranged into six chapters. In the first chapter we will discuss the literature review and past researches which have done in the strong field ionization. Moreover, we will briefly explain the motivations of research, objectives and the thesis outline in this chapter. Chapter 2 is devoted to the introduction of some basic theory and calculations that are involved in the strong field ionization. Besides, we will briefly introduce the other processes such as Above Threshold Ionization, Multiphoton Ionization and Corkum's simple man model to enable the readers to have a better understanding and perspective of the strong field physics.

In chapter 3, we explore the reasons of the breakdown of the perturbative theory and derive the theoretical formalism and calculation of the non-perturbative theory. By understanding the formalism of non-perturbative calculation, we will further explore the Keldysh model and spell out the formalism in details. The result of the photoionization rate will be discussed. Chapter 4 is concerning on generalize the Keldysh's model to adopt arbitrary momentum of photoelectron. The derivation of our exact model will be shown and further compared with the Keldysh's result.

Chapter 5 is devoted to the extension of Keldysh's formalism so that the initial state of the photoionization can be arbitrary energy level of the hydrogen wavefunction, meanwhile Keldysh only consider the hydrogen ground state wavefunction as the initial state.

Thus, we generalized the theoretical model for photoionization so that the pulse envelope function is included and the model can adopt arbitrary electric field strength of the laser field. Relativistic effect is taken consideration into our generalized model as the intensity of the laser field is getting higher. In the discussion, the photoionization results are plotted in angular distribution and comparison has been made on the relativistic and non-relativistic photoionization. We will show that the result can be retrieved back to Keldysh's result which is non-relativistic by reducing the intensity of the intense laser field.

The last chapter is where the thesis is concluded with a short recap and several thoughts of future works about photoionization. The works in this thesis have been published in well known ISI journals such as Phys. Rev. A and J. Opt. Soc. Am. B, as shown in Appendix A.

CHAPTER 2

THE FORMALISM OF LIGHT-MATTER INTERACTION

2.1 Introduction

The idea of intense laser field started from the first most relevant empirical finding in the development of intense-field science was the observation of breakdown of atomic gases in air when a laser was shone on it. In 1965, Voronov and Delone were using a ruby laser (Voronov & Delone, 1966) with photons of about 2eV which is much smaller than the ionization potentials of noble gas atoms Ar and Xe (>10eV) which served as targets. They reported not only the observation of ions but also a nonlinear dependence of the ions yields on the laser intensity. In the later work by Hall, Branscomb and Robinson, they observed the photo-detachment effect (Robinson & Geltman, 1967), in which a negative ion emitteed an electron by interaction with a beam of subthreshold laser photons. Following by many interesting findings which develope the intense laser field to become the pioneer study in the world.

In this chapter, the important formalism that involed in light-matter interaction (Federov, 1991) will be described in details. Those formalism such as gauge transformation, non-perturbative theory, atom field interaction in semi-classical approach (J.-M. Yuan & George, 1978), the perturbative formalism for strong field ionization are playing the important roles as the main engines that drive the development of the interaction between atom and intense laser field. The details study and derivation of these formalism will be shown for a better understanding to the readers.

The Hamiltonian is the main formalism in the atom field interaction and it is very important in order to describe an atom whose interaction with the electromagnetic field results from the fact that the nucleus is moving with instantaneous position $\alpha(t)$. (which is one of the four Hamiltonian where we will discuss in the following section.) This is the essential coordinate for a classical free electron when moving in the field without any interaction. In the next section, we will discuss the gauge transformation on four different Hamiltonians which is very useful in order to understand the photoionization models in the later part.

2.2 Gauge Transformation

The gauge transformation (Mittleman, 1993) is an important process for understanding each photoionization model in order to preserve the invariance of the equations. For instance, the following equation consists a phase transformation of the wavefunction,

$$\Psi \rightarrow \exp i\left(\sum_n \Upsilon_n\left(\mathbf{r}_n,t\right)\right)\Psi, \tag{2.1}$$

and a corresponding shift in the vector potential $\mathbf{A}_j\left(\mathbf{r}_j,t\right)$ and also the scalar potential V,

$$\mathbf{A}_j\left(\mathbf{r}_j,t\right) \rightarrow \mathbf{A}_j\left(\mathbf{r}_j,t\right) + \frac{\hbar c}{e_n}\nabla_n \Upsilon_n\left(\mathbf{r}_n,t\right), \tag{2.2}$$

$$V \rightarrow V - \frac{1}{\hbar}\sum_n \frac{d}{dt}\Upsilon_n\left(\mathbf{r}_n,t\right). \tag{2.3}$$

Hence, the Schrödinger equation remains unchanged. The constraint on the potentials and the Maxwell equations which are governing the field are unchanged provided that the Υ_n satisfies the wave equation as following

$$\left(\nabla^2 - \frac{1}{c^2}\frac{d^2}{dt^2}\right)\Upsilon_n\left(\mathbf{r}_n,t\right) = 0. \tag{2.4}$$

Gauge transformation is also known as a unitary transformation of the wavefunction and provide an altered form of the Hamiltonian which is very useful for the calculation in the later chapter. The Hamiltonian of the Schrödinger equation is defined as below,

$$H_1 = \sum_{j=1}^{Z}\frac{1}{2\mu}\left(\mathbf{p}_i + \frac{e}{c}\mathbf{A}\left(t\right)\right)^2 + \frac{1}{M_N}\sum_{i\neq j=1}^{Z}\left(\mathbf{p}_i + \frac{e}{c}\mathbf{A}\left(t\right)\right)\cdot\left(\mathbf{p}_j + \frac{e}{c}\mathbf{A}\left(t\right)\right) + V\left(\mathbf{X}_i \ldots \mathbf{X}_Z\right), \tag{2.5}$$

where μ is the reduced mass of the electron and $\mathbf{p}_i$ is the canonical momentum. The Schrödinger equation is defined as

$$\left(i\hbar\frac{d}{dt} - H_1\right)\Psi_1 = 0, \tag{2.6}$$

and the unitary transformation is applied on the wavefunction

$$\Psi_1 = \exp\left(-i\Phi_j\right)\Psi_j. \tag{2.7}$$

where the function Φ_j can be arbitrary function. The new wavefunction is having the same physical meaning as the old wavefunction. In other word, we can say that the wavefunction is invariant and it satisfies

$$\left(i\frac{d}{dt} - H_j\right)\Psi_j = 0, \tag{2.8}$$

where

$$H_j = e^{i\Phi_j} H_1 e^{-i\Phi_j} - \dot{\Phi}_j. \tag{2.9}$$

By setting Φ_j, it is obviously shown that the Hamiltonian remains unchanged.

However, the purpose of Φ_j is designed to eliminate the second order of the vector potential, A^2 from

$$\Phi_2 = \frac{Ze^2}{2mc^2}\frac{M_A}{M_N}\int^t dt' A^2\left(t'\right). \tag{2.10}$$

As a result, the Hamiltonian becomes

$$H_2 = \sum_{j=1}^{Z}\frac{p_j^2}{2\mu} + \sum_{i>j=1}^{Z}\frac{\mathbf{p}_i\cdot\mathbf{p}_j}{2M_N} + \frac{e}{mc}\frac{M_A}{M_N}\mathbf{A}\left(t\right)\times\sum_{j=1}^{Z}\mathbf{p}_j + V. \tag{2.11}$$

In general, the coupling to the field is only in the $\mathbf{p}\cdot\mathbf{A}$ term, nevertheless it can still contribute A^2 terns when the second order is taken into consideration. Meanwhile, we introduce another choice of Φ_j which will remove the $\mathbf{p}\cdot\mathbf{A}$ term

$$\Phi_3 = \frac{e}{c}\sum_{j=1}^{Z}\mathbf{X}_j\cdot\mathbf{A}\left(t\right), \tag{2.12}$$

and hence, the new Hamiltonian is obtained without the $\mathbf{p}\cdot\mathbf{A}$ term,

$$H_3 = \sum_{j=1}^{Z}\frac{p_j^2}{2\mu} + \sum_{i>j=1}^{Z}\frac{\mathbf{p}_i\cdot\mathbf{p}_j}{M_N} + e\mathbf{E}\left(t\right)\times\sum_{j=1}^{Z}\mathbf{X}_j + V, \tag{2.13}$$

where the classical electric field, $\mathbf{E}\left(t\right)$ is defined as

$$\mathbf{E}\left(t\right) = -\frac{d}{dt}\mathbf{A}\left(t\right). \tag{2.14}$$

Next, we would like to introduce an infinite possible number of Φ's which takes the form of

$$\Phi_4 = \frac{e}{mc}\frac{M_A}{M_N}\int_{-\infty}^{t} dt' A^2\left(t'\right) \times \sum_{j=1}^{Z} \mathbf{p}_j + \Phi_2. \tag{2.15}$$

In order to remives the $\mathbf{p}\cdot\mathbf{A}$ coupling, we introduce a transformation to the Kramers' gauge (refer to appendix D). The equation is expressed as

$$e^{i\Phi_4}\mathbf{X}_j e^{-i\Phi_4} = \mathbf{X}_j - \alpha\left(t\right), \tag{2.16}$$

where $\mathbf{f}\left(t\right)$ is defined as

$$\alpha\left(t\right) = \frac{e}{mc}\frac{M_A}{M_N}\int_{-\infty}^{t} dt' A\left(t\right), \tag{2.17}$$

and shows that this transformation is also can be known as a space translation or an acceleration transformation in the accelerated frame.

Hence, the new Hamiltonian is

$$H_4 = \sum_{j=1}^{Z}\frac{p_j^2}{2\mu} + \sum_{i>j=1}^{Z}\frac{\mathbf{p}_i\cdot\mathbf{p}_j}{M_N} + V\left(\mathbf{X}_1 - \alpha\left(t\right), \ldots\mathbf{X}_Z - \alpha\left(t\right)\right). \tag{2.18}$$

In other word, the above Hamiltonian as in Eq. 2.18 which is generated by Φ_4 in Eq. 2.15 has a physical meaning where the free electron is space translated to a frame which is unaccelerated, but the nucleus is still moving in the field itself.

The four Hamiltonian that we have discussed are all equally meaningful thus providing identical physical results when the Schrödinger equation is solved exactly. Gauge transformation (Brown & Kibble, 1964) may cause the result to be different if the gauge is not chosen wisely. Therefore, we shall understand that the different gauges may be useful in different system.

2.3 Interaction between Atom and Field

Atom field interaction (Claude Cohen-Tannoudji, 2012) involves coupling between the atom and the mode of the electromagnetic field. Description of the interaction is valid when the atomic levels involved are resonant with the driving field while other levels are highly detuned. The semiclassical theory treats the atom as a quantum three-level system and the radiation field classically. The three level atom undergoes optical Rabi oscillations (R. Ooi, Hazmin, & Singh, 2013) induced by the driving electromagnetic field. The oscillations experience damping as the atomic levels decay. A better understanding of this simple model of the atom-field interaction is crucial before engaging in more complex problems involving an ensembles of atoms interacting with the field such as laser. This also provides a platform for studying complicated atoms ensembles such as biomolecules.

The atom and field interaction can be described by two different theory which is the semiclassical theory and the quantum theory. The semiclassical theory predicts Rabi oscillations for atomic inversion ignoring decay process while the quantum theory predicts certain collapse and revival phenomena due to the quantum aspects of the field.

2.3.1 Semi-Classical Theory

Atom field interaction involves coupling between the atom and the mode of the electromagnetic field. Description of the interaction is valid when the atomic levels involved are resonant with the driving field while other levels are highly detuned (V. M. Akulin & Sartakov, 1977). The semiclassical theory treats the atom as a quantum three-level system and the radiation field classically. The three level atom undergoes optical Rabi oscillations induced by the driving electromagnetic field. The oscillations experience damping as the atomic levels decay. A better understanding of this simple model of the atom-field interaction is crucial before engaging in more complex problems involving an ensembles of atoms interacting with the field such as laser. This also provides a platform for studying complicated atoms ensembles such as biomolecules.

2.3.1 (a) Atom Field Interaction Hamiltonian

The minimal coupling Hamiltonian depicts about the interaction of an electron of charge e and mass m with an external electromagnetic field

$$H = \frac{1}{2m}\left[\mathbf{p} - e\mathbf{A}(\mathbf{r},t)\right]^2 + eU(\mathbf{r},t) + V(r), \tag{2.19}$$

where $\mathbf{p}$ is the canonical momentum operator, $\mathbf{A}(\mathbf{r},t)$ and $U(\mathbf{r},t)$ are the vector and scalar potentials of external electromagnetic field respectively and $V(r)$ is the electrostatic potential which acts as atomic binding potential. From here, we can see several approaches of the Hamiltonian.

1. Local Gauge (phase) Invariance and Minimal-Coupling Hamiltonian The motion of a free electron can be explained by the Schrodinger equation

$$\frac{-\hbar^2}{2m}\nabla^2\psi = i\hbar\frac{d\psi}{dt}. \tag{2.20}$$

The probability density for finding an electron at position **r** and time t is

$$P(\mathbf{r},t) = |\psi(\mathbf{r},t)|^2.$$

If the solution is $\psi(\mathbf{r},t)$, so does $\psi(\mathbf{r},t) = \psi(\mathbf{r},t)\exp(i\chi)$ where χ is an arbitrary constant phase and the probability density is unaffected by the choice of χ. This means two functions that differ by a constant phase factor still represents the same physical state. However, thing changes if phase is allowed to vary locally, by means to be a function of space and time variables such that $\psi(\mathbf{r},t) \to \psi(\mathbf{r},t)e^{i\chi(\mathbf{r},t)}$. Hence, the Scrodinger equation had to be modified to satisfy the local gauge invariance (Vanne & Saenz, 2009) by adding new terms to Eq. 3.114

$$\left\{\frac{-\hbar^2}{2m}\left[\nabla - i\frac{e}{\hbar}\mathbf{A}(\mathbf{r},t)\right]^2 + eU(\mathbf{r},t)\right\}\psi = i\hbar\frac{d\psi}{dt}, \tag{2.21}$$

where $\mathbf{A}(\mathbf{r},t) \to \mathbf{A}(\mathbf{r},t) + \frac{\hbar}{e}\nabla\chi(\mathbf{r},t)$ and $U(\mathbf{r},t) \to U(\mathbf{r},t) - \frac{\hbar}{e}\frac{d\chi}{dt}(\mathbf{r},t)$. The scalar and vector potnetial are gauge dependent potentials. The gauge-independent quantities are electric fields $\mathbf{E} = -\nabla U - \frac{d\mathbf{A}}{dt}$ and magnetic fields $\mathbf{B} = \nabla \times \mathbf{A}$. Take note that RHS of equation below is actually the Hamiltonian of Eq. 2.19. Thus, Scrodinger can be rewritten in terms of gauge-dependent quantities

$$\left\{\frac{1}{2m}\left[-i\hbar\nabla - e\mathbf{A}(\mathbf{r},t)\right]^2 + eU(\mathbf{r},t)\right\}\psi = i\hbar\frac{d\psi}{dt} \tag{2.22}$$

by using $\mathbf{p} = -i\hbar\nabla$. The electrons are described by wavefunction $\psi(\mathbf{r},t)$ whereas the field is depicted by the vector and scalar potentials A and U, respectively.

2. Dipole Approximation & $\mathbf{r}\cdot\mathbf{E}$ Hamiltonian Consider that the electron is bound by a potential $V(r)$ to the nucleus located at $\mathbf{r}_0$. Dipole approximation (Kylstra et al., 2000) simplifies the minimal-coupling Hamiltonian. The entire atom is immersed in a plane of electromagnetic wave depicted by a vector potential $\mathbf{A}(\mathbf{r}_0 + \mathbf{r},t)$ which is expressed in dipole approximation $\mathbf{k}\cdot\mathbf{r} << 1$ as

$$\begin{aligned}
\mathbf{A}(\mathbf{r}_0+\mathbf{r},t) &= \mathbf{A}(t)\exp(i\mathbf{k}\cdot(\mathbf{r}_0+\mathbf{r})) &(2.23)\\
&= \mathbf{A}(t)\exp(i\mathbf{k}\cdot\mathbf{r}_0)(1+i\mathbf{k}.\mathbf{r}+...) &(2.24)\\
&\simeq \mathbf{A}(t)\exp(i\mathbf{k}\cdot\mathbf{r}_0). &(2.25)
\end{aligned}$$

From Eq. 3.124, let $\mathbf{A}(\mathbf{r},t)=\mathbf{A}(\mathbf{r}_0,t)$ and binding potential $V(r)$ that denotes electrostatic potential that binds electron to nucleus, getting

$$\left\{\frac{-\hbar^2}{2m}\left[\nabla-i\frac{e}{\hbar}\mathbf{A}(\mathbf{r}_0,t)\right]^2+V(r)\right\}\psi(\mathbf{r},t)=i\hbar\frac{d\psi(\mathbf{r},t)}{dt}. \tag{2.26}$$

In the radiation gauge, $U(\mathbf{r},t)=0$ and $\nabla\cdot\mathbf{A}=0$. A new wave function $\phi(\mathbf{r},t)$ is defined which is in the form of

$$\psi(\mathbf{r},t)=\exp\left(i\frac{e}{\hbar}\mathbf{A}(\mathbf{r}_0,t)\cdot\mathbf{r}\right)\phi(\mathbf{r},t). \tag{2.27}$$

We pluck the Eq. 2.27 into Eq. 3.14 to get

$$\begin{aligned}
&\left\{\frac{-\hbar^2}{2m}\left[\nabla-i\frac{e}{\hbar}\mathbf{A}(\mathbf{r}_0,t)\right]^2+V(r)\right\}\exp\left(i\frac{e}{\hbar}\mathbf{A}(\mathbf{r}_0,t)\cdot\mathbf{r}\right)\phi(\mathbf{r},t)\\
&= i\hbar\frac{d}{dt}\exp\left(i\frac{e}{\hbar}\mathbf{A}(\mathbf{r}_0,t)\cdot\mathbf{r}\right)\phi(\mathbf{r},t), &(2.28)
\end{aligned}$$

then we reduce Eq. 2.28 into

$$\exp\left(i\frac{e}{\hbar}\mathbf{A}\cdot\mathbf{r}\right)\left[\frac{p^2}{2m}+V(r)\right]\phi(\mathbf{r},t)=i\hbar\left[\dot{\phi}(\mathbf{r},t)+i\frac{e}{\hbar}\dot{\mathbf{A}}\cdot\mathbf{r}\phi(\mathbf{r},t)\right]\exp\left(i\frac{e}{\hbar}\mathbf{A}\cdot\mathbf{r}\right), \tag{2.29}$$

and obtain the following expression

$$i\hbar\dot{\phi}(\mathbf{r},t)=\left[\left(\frac{p^2}{2m}+V(r)\right)+e\dot{\mathbf{A}}\cdot\mathbf{r}\right]\phi(\mathbf{r},t). \tag{2.30}$$

Next, we define unperturbed Hamiltonian of the electron as $H_0=\frac{p^2}{2m}+V(r)$ and use $E=-\dot{\mathbf{A}}$ yielding

$$i\hbar\dot{\phi}(\mathbf{r},t)=[H_0-e\mathbf{r}\cdot\mathbf{E}(\mathbf{r}_0,t)]\phi(\mathbf{r},t). \tag{2.31}$$

The total Hamiltonian is

$$H=H_0+H_1,$$

with

$$H_1 = -e\mathbf{r}\cdot\mathbf{E}(\mathbf{r}_0,t). \tag{2.32}$$

is in terms of gauge-independent field **E**.

3. $\mathbf{p}\cdot\mathbf{A}$ Hamiltonian Hamiltonian can also be expressed in terms of canonical momentum **p** and vector potential **A**. The radiation gauge (Bauer, Milosevic, & Becker, 2005) is followed where $U(\mathbf{r},t)=0$ and $\nabla\cdot\mathbf{A}=0$. In quantum mechanics, $[\mathbf{p},\mathbf{A}]=0$ due to $\nabla\cdot\mathbf{A}=0$. The total Hamiltonian is

$$H' = H_0 + H_2 \tag{2.33}$$

where $H_0=\frac{p^2}{2m}+V(r)$. In the dipole approximation of Eq. 2.25

$$H_2 = -\frac{e}{m}\mathbf{p}.\mathbf{A}(\mathbf{r}_0,t)+\frac{e^2}{2m}A^2(\mathbf{r}_0,t)$$

The Scrodinger equation becomes

$$H\psi(\mathbf{r},t) = i\hbar\frac{d}{dt}\psi(\mathbf{r},t) \tag{2.34}$$

$$\left[H_0-\frac{e}{m}\mathbf{p}.\mathbf{A}(\mathbf{r}_0,t)+\frac{e^2}{2m}A^2(\mathbf{r}_0,t)\right]\psi(\mathbf{r},t) = i\hbar\frac{d}{dt}\psi(\mathbf{r},t) \tag{2.35}$$

The A^2 term is usually small and is ignored. Thus

$$i\hbar\frac{d}{dt}\psi(\mathbf{r},t)=\left[H_0-\frac{e}{m}\mathbf{p}\cdot\mathbf{A}(\mathbf{r}_0,t)\right]\psi(\mathbf{r},t) \tag{2.36}$$

such that $H_2=-\frac{e}{m}\mathbf{p}\cdot\mathbf{A}(\mathbf{r}_0,t)$. H_1 and H_2 give different physical results because the matrix elements of these Hamiltonians calculated between the eigenstates of the unperturbed Hamiltonian H_0 are dissimilar. Next, we consider linearly polarized monochromatic plane wave field interacting with an atom placed at $r_0=0$,

$$E(0,t)=\widetilde{E}\cos\nu t \tag{2.37}$$

$$A(0,t)=-\frac{1}{\nu}\widetilde{E}\sin\nu t \tag{2.38}$$

and the time-independent amplitudes associated with H_1 and H_2

$$W_1=-e\mathbf{r}.\widetilde{E} \tag{2.39}$$

$$W_2 = \frac{e}{mv}\mathbf{p}.\widetilde{E} \tag{2.40}$$

The initial eigenstate $|i\rangle$ of $H_0 = H_0|i\rangle = \hbar\omega_i|i\rangle$ and a final eigenstate $|f\rangle, H_0|f\rangle = \hbar\omega_f|f\rangle$ with the frequency transformation, $\omega = \omega_f - \omega_i$.

2.4 Non-perturbative theory

For pedagogical reason, the wavefunction of a single particle moving in three dimensions can be described by the following equation,

$$i\hbar\Psi(r,t) = H(r,t)\Psi(r,t), \tag{2.41}$$

where the Hamiltonian (Tong & Chu, 1997) is defined as

$$H(r,t) = T_k + V(r,t) = -\frac{h^2}{2m}\nabla + V(r,t). \tag{2.42}$$

Before proceed to the Schrödinger equation, firstly we have to introduce the equation of continuity which is defined as

$$\rho(r,t) = -\nabla j(r,t), \tag{2.43}$$

with the so-called probability density

$$\rho(r,t) = |\Psi(r,t)|^2, \tag{2.44}$$

and the probability density flux which is described as the following expression

$$j(r,t) = \frac{\hbar}{m}Im\{(r,t)\} = -\frac{1}{m}Re\{\Psi^*(r,t)p\Psi(r,t)\}. \tag{2.45}$$

We refer to the above equation 2.45, both LHS and RHS of this equation must be equal to a constant, which we name as E. Hence, we thus arrive at the two equations,

$$i\hbar\dot{\varphi}(t) = E\varphi(t), \tag{2.46}$$

$$H(r)\psi_e(r) = E\psi_e(r). \tag{2.47}$$

The first of these equations, equation 2.46 can be solved immediately and yields,

$$\varphi(t) = \varphi_0 e^{-iEt/h}. \tag{2.48}$$

Next, the solution for time-dependent Schrödinger equation is obtained as following,

$$\Psi(r,t) = \psi E(r)\varphi_0 e^{iEt/h}. \tag{2.49}$$

We would rewrite a general solution of the time-dependent Schrödinger equation which is a linear combination of eigenfunctions,

$$\Psi(r,t) = \sum_{n=0}^{\infty} a_n \psi_n(r) e^{-iEt/h} \tag{2.50}$$

or another expression,

$$\Psi(r,t) = \int dEa(E)\psi_e(r)e^{iEt/h} \tag{2.51}$$

Next, we shall rewrite the time-dependent schrodinger equation 2.41 into the operator form and ket, which is

$$i\hbar \big|\Psi(t)\rangle = \hat{H}\big|\Psi(t)\rangle. \tag{2.52}$$

A formal solution of this equation is given by

$$\Big|\Psi(t)\rangle = e^{iH(t-t0)/h}\Big|\Psi(t_0)\rangle = U(t,t_0)\,|\Psi(t_0)|\rangle, \tag{2.53}$$

and it can be shown that the integral as following expression

$$\langle\Psi(t)\Psi(t)\rangle = \Big\langle\Psi(t_0)\Big|e^{i\hat{H}t(-t_0)/h}e^{-iH(t-t_0)/h}\Big|\Psi(t_0)\Big\rangle = \langle\Psi(t_0)\Psi(t_0)\rangle\,, \tag{2.54}$$

which is equivalent to the time-evolution operator being unitary

$$U(t,t_0) = U^{-1}(t,t_0). \tag{2.55}$$

Let us recall the composition property of the time-evolution operator (Bandrauk, 1994)

$$U(t,t_0) = U(t,t')U(t',t_0). \tag{2.56}$$

Hence, the wavefunction can be shown by insertion

$$|\Psi(t)\rangle = |\Psi(t_0)\rangle - \frac{1}{h}\int_{t0} dt'\hat{H}(t')\Psi(t'). \tag{2.57}$$

On the contrary, let us introduce a special wavefunction, the closure relation, which is defined as

$$K(r,t;r'0) = \int d^3r'' K(r,t;r'',t')K(r'',t'';r',0), \tag{2.58}$$

and also the Feynman-Kac formula (Moral, 2004),

$$E_0 \quad = \quad -\lim -InG(-ihr,0), \tag{2.59}$$

$$\check{T}\left[\breve{A}(t_1)B(t_2)\right] \quad \equiv \quad \left\{\frac{B(t_2)A(t_1)}{A(t_1)B(t_2)}\right\}. \tag{2.60}$$

Therefore, the time-ordering operator can be shown as

$$\breve{U}(t,t_0) = \check{T}e^{-i/h}\int^t dt'\hat{H}(t'). \tag{2.61}$$

As a result, the propagated wavefunction becomes

$$\Psi(r,t) = \int d^3r' \left\langle r|U(t,0)|r'\right\rangle \Psi(r',0), \tag{2.62}$$

After replacing the time-ordering operator into the closure relation, hence the position matrix element of the time-evolution operator would be

$$K(r,t;r',0) = \left\langle r|U(t,0)|r'\right\rangle. \tag{2.63}$$

Let us define another special wavefunction, the closure relation which is defined as

$$K(r,t;r',0) = \int d^3r'' K(r,t;r'',t'')K(r'',t'';r',0), \tag{2.64}$$

thus, at the spectral representation, it can be rewritten as

$$K(r,t;,r',0)=\sum_{n=0}^{\infty}\Psi_n^*(r')\Psi_n(r). \tag{2.65}$$

By considering the auto-correlation function of an initial wavefunction, therefore,

$$\Psi\alpha=\sum_n|n\rangle\langle n|\sum_n cn, \tag{2.66}$$

which is defined according to

$$C\alpha\alpha(t)=\left\langle\Psi\alpha\left|e^{-iHt/h}\right|\Psi\alpha\right\rangle=\sum_n|C_n|^2e^{-iEn^{t/h}}. \tag{2.67}$$

Hence, one gains the local spectrum by Fourier transformation

$$\begin{aligned} S(\omega) &= \frac{1}{2\pi h}\int dte^{iwt}\alpha\alpha(t) \\ &= \sum_{n=0}^{\infty}|C_n|^2\delta(E_{n^{-h\omega}}). \end{aligned} \tag{2.68}$$

To this end, we perform the time evolution on equation 2.67 and yield,

$$\Psi\alpha(-ih\tau)=\sum_{n_0}C_n|n\rangle e^{-rEn}, \tag{2.69}$$

and we perform the Taylor expansion of the potential around $=qt$ accoding to

$$V(,t)V(qt,t)+V'(qt,t)(-qt)+\frac{1}{2!}V''(qt,t)(-qt)^2. \tag{2.70}$$

By using the time-dependent Schrödinger equation again, after the insertion of the time and position derivatives of the wavefunction, we can get the first and second derivative of the wavefunction as the following expression,

$$\begin{aligned} \Psi(x,t) &= -\alpha_t(x-qt)^2+2\alpha_tq_t(x-q_t)+\frac{i}{h}pt(x-qt)-\frac{i}{h}p_tq_t+\frac{i}{h}\delta t\}, \quad (2.71) \\ \Psi'(r,t) &= [-2\alpha_t(x-qt)+\frac{i}{h}pt]^2\}\Psi(x,t), \quad (2.72) \\ \Psi''(x,t) &= \{-2\alpha_t+[-2\alpha_t(x-qt)+\frac{i}{h}pt]^2\}\Psi(x,t). \quad (2.73) \end{aligned}$$

The wavefunction along the straight lines originating from $i=0$ can thus be written as

$$\begin{aligned}\Psi(2k\tau,\tau) &= \sqrt{\frac{2}{LN}\frac{1}{2i}q^{-(k/2)^2}}\sum_{n=1}^{N}\{q^{(n+k/2)^2}-q^{(n-k/2)^2}\}, \\ & \sqrt{\frac{2}{LN}\frac{1}{2i}q^{-(k/2)^2}}.\end{aligned} \tag{2.74}$$

To this end a simplified version of the short-time propagator with a simple end point rule for the discretization of the potential part of the action by replacing

$$V(\frac{xj+xj-1}{2}), \tag{2.75}$$

with

$$V(xj-1). \tag{2.76}$$

By defining the deviation from the classical path as

$$\eta(t') = x(t') - x_{cl}(t'), \tag{2.77}$$

the second-order expansion needed for the SPA is given by

$$S[x] = S[x_{cl}] + \frac{1}{2}\int dt'\eta(t')\breve{O}\eta(t'), \tag{2.78}$$

with

$$\breve{O} = -m\frac{d^2}{dt^2} - V''. \tag{2.79}$$

From basic classical mechanics we have the identity which is defined as

$$\frac{\partial^2 S[x_{cl}}{\partial x_f \partial x_i} = -\frac{\partial p_i}{\partial x_f}. \tag{2.80}$$

Hence, we have the Schrödinger equation in the interaction picture, where the perturbation Hamiltonian in the interaction picture is given by

$$\hat{W}_1(t,0) := \breve{U}_0(t,0)\hat{W}(t)\breve{U}_0(t,0). \tag{2.81}$$

Thus, the total Hamiltonian shall be of the form

$$\hat{H} = \sum_{n-1}^{2} \hat{H}_n(x_n) + V_{12}(x_n), \tag{2.82}$$

with single particle operators

$$\hat{H}_n(x_n) = -\frac{h^2}{2m}\frac{\partial^2}{\partial x_n^2} + V_n(x_n), \tag{2.83}$$

and the coupling potential V_{12} depending on the two coordinates in a non additive manner. The so-called Hartree Ansatz (Caillat et al., 2005) for the wavefunction is of the form

$$\Psi(x_{1,}x_{2,}t) = \Psi_1(x_{1,}t)\Psi_2(x_{2,}t). \tag{2.84}$$

This Ansatz is exact in the case that the coupling V_{12} vanishes, the single particle functions then fulfill

$$ih\Psi_n(x_n,t) = \hat{H}_n\Psi_n(x_{n,}t). \tag{2.85}$$

We now plug the Hartree Ansatz into the full time-dependent Schrödinger equation and find

$$ih(\Psi_2\Psi_1 + \Psi_1\Psi_2) = \Psi_2\hat{H}_1\Psi_1 + \Psi_1\hat{H}_2\Psi_2 + V_{12}\Psi_1\Psi_2. \tag{2.86}$$

By using the single particle equations of zeroth order with the index 2, the second terms on the LHS and the RHS cancel each other and one finds

$$ih\Psi_1(x_{1,}t) = \left(-\frac{h^2}{2m}\triangle_1 + V_{1,off}(x_{1,}t)\right)\Psi_1(x_1,t) \tag{2.87}$$

with an effective, time-dependent potential

$$V_{1,off}(r_{1,}t) = V_1(x_1) + (\Psi_2\,|V_{12}|\,\Psi_2)_2. \tag{2.88}$$

An analogous equation can be derived for particle 2

$$ih\Psi_2(x_{2,}t) = \left(-\frac{h^2}{2m}\triangle_2 + V_{2,off}(x_{2,}t)\right)\Psi_2(x_{2,}t), \tag{2.89}$$

and thus having the full solution of the time-independent Schrödinger equation

$$\hat{H}\psi_n(x,X) = E_n\psi_n(x,X), \tag{2.90}$$

with the Hamiltonian

$$\hat{H} = \frac{\widehat{p}^2}{2m} + \frac{\widehat{p}^2}{2M} + v(r,X) + V(X). \tag{2.91}$$

2.5 Time-Dependent Quantum Theory

Time-dependent quantum theory(Kulander, 1988) is the main engine that drives the non-perturbative theory, we shall start with the Hamiltonian as the following

$$\hat{H}^0(x\left|X)\phi_{j(x}\right|X) = \varepsilon_j^0(X)\phi_j(xX), \tag{2.92}$$

where

$$\hat{H}^0(xX) = \frac{\widehat{p}^2}{2m} + v(x,X), \tag{2.93}$$

depends parametrically on X and j is the quantum number of the light particle. By using the product Ansatz (Reiss, 2008), we have

$$\psi_n(x,X)\phi_j(xX)x_{1,j}(X), \tag{2.94}$$

and one arrives at equations of the form

$$\hat{H}_j^1(X)x1,j(X) = \varepsilon_l^1 x1,j(X), \tag{2.95}$$

with the Hamltonian

$$\hat{H}_j^1(X) = \frac{\widehat{p}^2}{2M} + V(X) + \varepsilon_j^0(X). \tag{2.96}$$

As a result, this yields coupled differential equations for the coefficients

$$ihc_j(t) = \varepsilon_j^0 c_j - ihX\sum_k d_{j,}k^c k. \tag{2.97}$$

In dealing with laser driven systems the problem of time-periodic Hamiltonians is of central importance. However in this case, we have

$$\hat{H}(t+T) = \hat{H}(t). \tag{2.98}$$

In order to solve the time-dependent Schrödinger equation, we prove that the Hamiltonian is extended by the time derivative

$$\hat{H}(t) \equiv \hat{H}(t) - ih\theta_t, \tag{2.99}$$

and the time-derivative of the exponential part yields

$$\hat{H}(x,t)\psi_\alpha(r,t) = \varepsilon_\alpha \psi_\alpha(r,t). \tag{2.100}$$

Hence, the wavefunction is written as a superposition of quasi-eigenfunctions

$$\Psi(t) = \sum_{\alpha'} c_{\alpha'} \psi_{\alpha'}(t) \exp\left\{-\frac{i}{h}\varepsilon_{\alpha'} t\right\}, \tag{2.101}$$

with appropriate coefficients

$$c_{a'} = \langle \psi_{\alpha'}(0)\Psi(0)\rangle. \tag{2.102}$$

The eigenfunctions of a certain (simple) Hamiltonian, as an example, the harmonic oscillator as the following expression

$$\hat{H}_{HO} = -\frac{h^2}{2m}\frac{d^2}{dx^2} + \frac{1}{2}m\omega_e^2 x^2. \tag{2.103}$$

The alternative representation of the harmonic oscillator Hamiltonian

$$\hat{H}_{HO} = h\omega_e\left(\breve{a}'\breve{a} + \frac{1}{2}\right). \tag{2.104}$$

An arbitrary time-dependent wavefunction can now be expanded into eigenfunctions of the harmonic oscillator according to

$$\Psi(t) = \sum_{l=0}^{\infty} d_l(t) l. \tag{2.105}$$

Due to the periodic timedependence of the Floquet functions they can be Fourier expanded according to

$$|\psi_\alpha(t)\rangle = \sum_{n=-\infty}^{\infty} |\psi_a^n\rangle e^{in\omega t}. \tag{2.106}$$

The Fourier coefficients on the RHS of equation 2.106 can in turn be expanded in an orthogonal system $\{|k\}$

$$|\psi_a^n\rangle = \sum_{k=0}^{\infty} \psi_{k,\alpha}^n |k\rangle, \tag{2.107}$$

and the time integration yields

$$\hat{H}^{[m-n]} = \hat{H}_0 \delta_{m,n} + \frac{\hat{H}_1}{2i}\{\delta_{mn-1} - \delta_{mn+1}\}. \tag{2.108}$$

Then, this leads to the fact that the exponentiated operator of kinetic energy becomes local and can be applied easily via

$$\begin{aligned} \left\langle x'' \left| e^{-i\tau_k \triangle t/h} \right| p' \right\rangle &= \left\langle x'' p' \right\rangle e^{\tau_{k(p')\triangle t/h}} \\ &= \frac{1}{\sqrt{2\pi h}} e^{ip'r''/h} e^{iTk(p')\triangle t/h}. \end{aligned} \tag{2.109}$$

The discrete version of the Fourier transform is

$$\Phi(x_i) = \sum_{k=-N/2-1}^{N/2} a_k e^{2\pi i k r_1/X}, \tag{2.110}$$

$$a_k = \frac{1}{N}\sum_{n=1}^{N} \Phi(x_n) e^{-2\pi i k x_n/X}. \tag{2.111}$$

The maximal momentum that can be described is

$$p_{\max} = h/(2\triangle x) = Nh/(2X). \tag{2.112}$$

This can be avoided by adding a negative imaginary potential of the form

$$V(x) = -if(x)\Theta(x - x_a). \tag{2.113}$$

An at least conditionally stable method can be constructed by application of the second-order formula

$$\Psi(t)\frac{\Psi(t+\triangle t)-\Psi(t-\triangle t)}{2\triangle t}. \tag{2.114}$$

The condition under which it is stable can be derived by considering the eigenvalues of the propagation matrix that appears by using the discrete form of the time-derivative

$$\left(\frac{\Psi_{n+1}}{\Psi_n}\right)=\left(\frac{1-4\hat{H}^2\triangle t^2/h^2-2i\hat{H}\triangle t/h}{-2i\hat{H}\triangle t/h}\right)\left(\frac{\Psi_{n-1}}{\Psi_{n-2}}\right). \tag{2.115}$$

By replacing the operator $\hat{H}$ with E, the eigenvalues of the matrix are

$$\lambda_{1,2}=1-2E^2\triangle t^2/h^2\pm\frac{2E\triangle t}{h}\sqrt{\frac{E^2\triangle t^2}{h^2}}-1, \tag{2.116}$$

and the first-order formula is

$$\breve{U}(\triangle t)\widehat{1}-i\hat{H}\triangle t/h. \tag{2.117}$$

Hence, equating the gained expressions yields

$$(\widehat{1}+i\hat{H}\triangle t/h)|\Psi_{n+1}\rangle=(\widehat{1}-i\hat{H}\triangle t/h)|\Psi_{n-1}\rangle. \tag{2.118}$$

Due to its implicit nature the method requires a matrix inversion and formally leads to the prescription (also referred to as Cayley approximation)

$$|\Psi_n\rangle=\frac{\widehat{1}-i\hat{H}\triangle t/(2h)}{\widehat{1}+i\hat{H}\triangle t/(2h)}|\Psi_{n-1}\rangle. \tag{2.119}$$

The idea behind polynomial methods is the expansion of the time-evolution operator in terms of polynomials, according to

$$e^{-iHt/h}=\sum_n a_n p_n(\hat{H}). \tag{2.120}$$

Formally, the equation above can be integrated over a small time step, yielding

$$\eta(t+\triangle t)=\exp\{-\triangle t\hat{H}\}\eta(t). \tag{2.121}$$

The basis for the reformulation of the semiclassical propagator is the matrix element of the time-evolution operator 1 and 3 between coherent states

$$K(zf,t;z_i;0) \equiv \left\langle z_f \left| e^{-i\hat{H}t} \right| z_i \right\rangle, \tag{2.122}$$

which is known as the Herman–Kluk Propagator. This procedure yields

$$K^{HK}(x_f,t;x_i,0 \equiv \int \frac{d^N p_i d^N q_i}{(2\pi h)^N} \langle x_f |\check{z}_t|\rangle R(p_i,q_i,t) \exp\{\frac{i}{h}S(p_i,q_i,t)\} \langle \check{z}_i |x_i|\rangle . \tag{2.123}$$

Definitions that are used in the expression above are the classical action functional, that depends on the initial phase space variables and for this reason is written (and denoted) as a function here according to

$$S(p_i,q_i,t) \equiv_0^t dt'[p_{t'}q_{t'} - H]. \tag{2.124}$$

Furthermore,

$$R(p_i,q_i,t) \equiv \left| \frac{1}{2} \left(m_{11} + m_{22} - ih\gamma m_{21} - \frac{1}{ih\gamma} m_{12} \right) \right|^{1/2}, \tag{2.125}$$

and the mixed matrix element

$$K(x_f,t;\check{z}_\alpha,0) \equiv \left\langle x_f \left| e^{-i\hat{H}t/} \right| \check{z}_\alpha \right\rangle = \int d^N x_i K(x_f,t;,x_i,0) \langle x_i |\check{z}\alpha|\rangle , \tag{2.126}$$

with the time-dependent $N \times N$ width parameter matrix

$$\gamma t = \gamma(m_{11} + \frac{1}{i\gamma h} m_{12})(m_{22} + i\gamma h m_{21})^{-1}, \tag{2.127}$$

with the width parameter which is

$$\alpha_t = \alpha_0 \frac{m_{11} + \frac{1}{2i\alpha_0 h} m_{12}}{m_{22} + 2i\alpha_0 h m_{21}}. \tag{2.128}$$

The displayed quantity is the auto-correlation function

$$c(t) \equiv \langle \Psi(0)\Psi(t) \rangle . \tag{2.129}$$

As a result, we obtain the Morse potential with dimensionless Hamiltonian

$$\hat{H} = \frac{\widehat{p}}{2} + D(1 - \exp\{-\lambda x\})^2. \tag{2.130}$$

Nevertheless, we introduce the Weyl transformation which is defined as the following

$$A(p,q) = \int du e^{iqu/h} \left\langle p + u/2 \left| \breve{A} \right| p - u/2 \right\rangle, \tag{2.131}$$

and the inverse transformation is

$$\begin{aligned} \breve{A} &= \frac{1}{h} \int dp dq A(p,q) \widehat{\triangle}(p,q), &(2.132)\\ \widehat{\triangle} &= \int dv e^{ipv/h} \left| q + v/2 >< q - v/2 \right|. &(2.133) \end{aligned}$$

Hence, we apply Weyl transformation on the Hamiltonian and yield,

$$H(p,q) = \frac{p^2}{2m} + V(q). \tag{2.134}$$

In general, the variation of a functional is defined via

$$\delta\Phi \equiv \Phi[h] = \int dx \frac{\delta\Phi}{\delta h(x)} \delta h(x). \tag{2.135}$$

For the specific cases which are

$$\begin{aligned} \Phi_1[h] &= \int dx h(x) f(x), &(2.136)\\ \Phi_2[h] &= \int dx F(x, h(x)), &(2.137)\\ \Phi_3[h] &= \int dx F\left(x, h(x), \frac{dh(x)}{dx}\right). &(2.138) \end{aligned}$$

After perform the second variation, we obtain the following expression

$$\begin{aligned} \delta^2 S[x_{cl}] &= \delta S[x_{cl} + \eta] - \delta S[x_{cl}] \int dt' \{-m \frac{d^2}{dt^2}(x_{cl} + \eta) - V'(x_{cl} + \eta)\}\eta - \int dt' \{-m x_{cl} - V'(x_{cl})\} \\ &= \int dt' \{-m\eta - V''(x_{cl})\eta\}\eta \\ &= \int dt' \eta \breve{O} \eta. &(2.13 \end{aligned}$$

The determinant as defined as $det(M) = det(m22) \times det(m11m12 - m12m21)$ is only valid for block matrices which is

$$\delta[x_f - q_t(p_{i,X_I})] = \sum_j \frac{1}{\|\partial q_t/\partial p_i\|}\delta(p_i - p_j). \tag{2.140}$$

2.6 Light Matter Interaction in Weak Field

The Hamiltonian in the dipole approximation depicts the interaction of a radiation field $\mathbf{E}$ with a single electron atom which is in the form of

$$H = H_A + H_F - e\mathbf{r}\cdot\mathbf{E} \tag{2.141}$$

H_A and H_F are the energies of the atom and the radiation field, respectively, without interaction and $\mathbf{r}$ is the position vector of the electron. The field is assumed to be uniform over the whole atom for dipole approximation.

The energy of free field H_F in terms of creation and destruction operators is defined as

$$H_F = \sum_{\mathbf{k}} \hbar \nu_k \left(a_{\mathbf{k}}^{+} a_{\mathbf{k}} + \frac{1}{2}\right) \tag{2.142}$$

The atom transition operators is $\sigma_{ij} = |i\rangle\langle j|$. Since $\{|i\rangle\}$ represents a complete set of atomic energy eigenstates as $\sum_i |i\rangle\langle i| = 1$. The eigenvalue equation $H_A|i\rangle = E_i|i\rangle$ equals to

$$H_A = \sum_i E_i |i\rangle\langle i| = \sum_i E_i \sigma_{ii} \tag{2.143}$$

The final term of Hamiltonian is

$$e\mathbf{r} = \sum_{i,j} e|i\rangle\langle i|\mathbf{r}|j\rangle\langle j| = \sum_{i,j} e\langle i|\mathbf{r}|j\rangle|i\rangle\langle j| = \sum_{i,j} \wp_{ij}\sigma_{ij} \tag{2.144}$$

where $\wp_{ij} = e\langle i|\mathbf{r}|j\rangle$ is the electric-dipole transition matrix element. Assume $\wp_{ij}$ to be real.

The electric field operator is determined in the dipole approximation at the position of the point atom. Considering atom at origin

$$\mathbf{E} = \sum_{\mathbf{k}} \hat{\varepsilon}_{\mathbf{k}} \xi_{\mathbf{k}} \left(a_{\mathbf{k}} + a_{\mathbf{k}}^{+}\right) \tag{2.145}$$

where $\xi_{\mathbf{k}} = (\hbar v_k / 2\varepsilon_0 V)^{1/2}$ (Scully, 1997). Consider a linear polarization basis and real polarization unit vectors for simplification.

2.6.0 (b) Interaction of a Single Atom with a Single-Mode Field

The Hamiltonian elucidates the interaction of single mode (Pegg & Barnett, 1989) quantized field of frequency v with a single 2 level atom which is in the form of

$$H = H_0 + H_1, \tag{2.146}$$

where

$$H_0 = \hbar v a^+ a + \frac{1}{2}\hbar\omega\sigma_z, \tag{2.147}$$

$$H_1 = \hbar g\left(\sigma_+ a + a^+ \sigma_-\right). \tag{2.148}$$

The atom-field interaction is explained in the dipole and rotating-wave approximations. In the interaction picture, the Hamiltonian is in the form of

$$\begin{aligned} V &= e^{iH_0 t/\hbar} H_1 e^{-iH_0 t/\hbar} && (2.149) \\ &= e^{i\left(va^+a + \frac{1}{2}\omega\sigma_z\right)t} \hbar g\left(\sigma_+ a + a^+\sigma_-\right) e^{-i\left(va^+a + \frac{1}{2}\omega\sigma_z\right)t}. && (2.150) \end{aligned}$$

The mathematical relation $e^{\alpha A} B e^{-\alpha A} = B + \alpha\,[A,B] + \frac{\alpha^2}{2!}\,[A,[A,B]] + ..$ is used to get $e^{iva^+at} a e^{-iva^+at} = a e^{-ivt}$ and $e^{i\omega\sigma_z t/2}\sigma_+ e^{-i\omega\sigma_z t/2} = \sigma_+ e^{i\omega t}$.

$$\begin{aligned} e^{iva^+at} a e^{-iva^+at} &= a + ivat\left[a^+, a\right] + \frac{(ivat)^2}{2!}\left[a^+, \left[a^+, a\right]\right] + ... \\ &= a\left(1 - ivt + \frac{(ivt)^2}{2!}\right) \\ &= a e^{-ivt}, && (2.151) \end{aligned}$$

and the second relation yields

$$\begin{aligned} e^{i\omega\sigma_z t/2}\sigma_+ e^{-i\omega\sigma_z t/2} &= \sigma_+ + i\omega t/2\,[\sigma_z, \sigma_+] \\ &= \sigma_+ (1 + i\omega t) \\ &= \sigma_+ e^{i\omega t}. && (2.152) \end{aligned}$$

Then, for the creatian operator, we have

$$\begin{aligned} e^{iva^+at}a^+e^{-iva^+at} &= a^+ + iva^+t\left[a,a^+\right] \\ &= a^+\left(1+ivt\right) \\ &= a^+e^{ivt}, \end{aligned} \quad (2.153)$$

and

$$\begin{aligned} e^{i\omega\sigma_z t/2}\sigma_- e^{-i\omega\sigma_z t/2} &= \sigma_- + \frac{i\omega t}{2}\left[\sigma_z,\sigma_-\right] \\ &= \sigma_-\left(1-i\omega t\right) \\ &= \sigma_- e^{-i\omega t}. \end{aligned} \quad (2.154)$$

as $[\sigma_z,\sigma_+] = 2\sigma_+$ and $[\sigma_z,\sigma_-] = -2\sigma_-$.

All the above expressions are then pluck into the relations to get Hamiltonian

$$\begin{aligned} V &= e^{i\left(va^+a+\frac{1}{2}\omega\sigma_z\right)t}\hbar g\left(\sigma_+a+a^+\sigma_-\right)e^{-i\left(va^+a+\frac{1}{2}\omega\sigma_z\right)t} \\ &= \hbar g\left[e^{iva^+at}ae^{-iva^+at}e^{\frac{i\omega\sigma_z t}{2}}\sigma_+e^{-\frac{i\omega\sigma_z t}{2}} + e^{iva^+at}a^+e^{-iva^+at}e^{\frac{i\omega\sigma_z t}{2}}\sigma_-e^{-\frac{i\omega\sigma_z t}{2}}\right] \\ &= \hbar g\left[\sigma_+ae^{i(\omega-v)t} + a^+\sigma_-e^{i(v-\omega)t}\right]. \end{aligned} \quad (2.155)$$

The detuning is defined as $\Delta = \omega - v$ yielding

$$V = \hbar g\left(\sigma_+ae^{i\Delta t} + a^+\sigma_-e^{-i\Delta t}\right). \quad (2.156)$$

The are three equivalent methods to solve for the evolution of the atom-field system expressed by Hamiltonian which are probability amplitude method, Heisenberg operator method and unitary time-evolution operator method.

2.6.0 (c) Interaction of a single two-level atom with a single-mode field

1. Probability Amplitude method Consider the interaction of a single-mode radiation field (Glauber, 1963) of frequency v with a *2* level atom. $|a\rangle$ and $|b\rangle$ indicates the upper level and lower level states of the atom where they are eigenstates of the unperturbed part of Hamiltonian H_0 with the eigenvalues $\hbar\omega_a$ and $\hbar\omega_b$ respectively. The wavefunction of the *two*-level atom can be expressed in the form of

$$|\psi(t)\rangle = C_a(t)|a\rangle + C_b(t)|b\rangle, \tag{2.157}$$

where C_a and C_b are the probability amplitudes of finding the atom in states $|a\rangle$ and $|b\rangle$, respectively. The Scrodinger equation is

$$|\dot{\psi}(t)\rangle = -\frac{i}{\hbar}H|\psi(t)\rangle, \tag{2.158}$$

$H = H_0 + H_1$ where H_0 is the unperturbed Hamiltonian and H_1 represents the interaction parts of the Hamiltonian. The completeness relation of $|a\rangle\langle a| + |b\rangle\langle b| = 1$ is used to write H_0 as

$$\begin{aligned} H_0 &= (|a\rangle\langle a| + |b\rangle\langle b|) H_0 (|a\rangle\langle a| + |b\rangle\langle b|) && (2.159)\\ &= (H_0|a\rangle\langle a||a\rangle\langle a| + H_0|b\rangle\langle b||b\rangle\langle b|) \\ &= \hbar\omega_a|a\rangle\langle a| + \hbar\omega_b|b\rangle\langle b|, && (2.160)\end{aligned}$$

where $H_0|a\rangle = \hbar\omega_a|a\rangle$ and $H_0|b\rangle = \hbar\omega_b|b\rangle$.

As for interaction of atom and radiation field, H_1 is rewritten as

$$\begin{aligned} H_1 &= -exE(t) \\ &= -e(|a\rangle\langle a| + |b\rangle\langle b|)x(|a\rangle\langle a| + |b\rangle\langle b|)E(z,t) \\ &= -e(\langle a|x|b\rangle|a\rangle\langle b| + \langle b|x|a\rangle|b\rangle\langle a|)E(z,t). && (2.161)\end{aligned}$$

The matrix element of the electric dipole moment$\wp_{ab} = e\langle a|x|b\rangle = \wp_{ba}^*$ and $E(t)$ is the electric field at the atom. Thus

$$H_1 = -(\wp_{ab}|a\rangle\langle b| + \wp_{ba}|b\rangle\langle a|)E(t). \tag{2.162}$$

The electric field is assumed to be linearly polarized along the x-axis as expressed as $E(t) = \widetilde{E}\cos\nu t$ in dipole approximation where E is the amplitude and $\nu = ck$ is the frequency of the field. Pluck in relevant equations into Scrodinger equation

$$\begin{aligned} |\dot{\psi}(t)\rangle &= -\frac{i}{\hbar}H|\psi(t)\rangle && (2.163)\\ \dot{C}_a(t)|a\rangle + \dot{C}_b(t)|b\rangle &= -\frac{i}{\hbar}\{(\hbar\omega_a|a\rangle\langle a| + \hbar\omega_b|b\rangle\langle b| && (2.164)\\ &\quad -(\wp_{ab}|a\rangle\langle b| + \wp_{ba}|b\rangle\langle a|)E(t))[C_a(t)|a\rangle + C_b(t)|b\rangle]\}, \end{aligned}$$

and hence

$$\begin{aligned}\dot{C}_a|a\rangle+\dot{C}_b|b\rangle &= -\frac{i}{\hbar}[\hbar\omega_a C_a|a\rangle\langle a||a\rangle+\hbar\omega_a C_b|a\rangle\langle a||b\rangle+\hbar\omega_b C_a|b\rangle\langle b||a\rangle \quad (2.165)\\ &+\hbar\omega_b C_b|b\rangle\langle b||b\rangle-\wp_{ab}EC_a|a\rangle\langle b||a\rangle-\wp_{ab}EC_b|a\rangle\langle b||b\rangle\\ &-\wp_{ba}EC_a|b\rangle\langle a||a\rangle-\wp_{ba}EC_b|b\rangle\langle a||b\rangle].\end{aligned}$$

Then we substitute $E(t)=\widetilde{E}\cos\nu t$

$$\dot{C}_a|a\rangle+\dot{C}_b|b\rangle = -\frac{i}{\hbar}[\hbar\omega_a C_a|a\rangle+\hbar\omega_b C_b|b\rangle-\wp_{ab}\widetilde{E}\cos\nu t C_b|a\rangle \quad (2.166)$$

$$-\wp_{ba}\widetilde{E}\cos\nu t C_a|b\rangle], \quad (2.167)$$

and multiply with $\langle a|$

$$\begin{aligned}\dot{C}_a\langle a||a\rangle+\dot{C}_b\langle a||b\rangle &= -\frac{i}{\hbar}[\hbar\omega_a C_a\langle a||a\rangle+\hbar\omega_b C_b\langle a||b\rangle \quad (2.168)\\ &-\wp_{ab}\widetilde{E}\cos\nu t C_b\langle a||a\rangle-\wp_{ba}\widetilde{E}\cos\nu t C_a\langle a||b\rangle],\end{aligned}$$

with

$$\dot{C}_a=-i\omega_a C_a+i\frac{\wp_{ab}\widetilde{E}}{\hbar}\cos(\nu t)C_b. \quad (2.169)$$

The Rabi frequency is defined as $\Omega_R=\frac{|\wp_{ba}|\widetilde{E}}{\hbar}$ and ϕ is the phase of the dipole matrix element $\wp_{ba}=|\wp_{ba}|\exp(i\phi)$. Hence, the equation of motion for amplitude C_a is

$$\begin{aligned}\dot{C}_a &= -i\omega_a C_a+i\frac{|\wp_{ab}|\widetilde{E}}{\hbar}\exp(-i\phi)\cos(\nu t)C_b\\ &= -i\omega_a C_a+i\Omega_R e^{-i\phi}\cos(\nu t)C_b, \quad (2.170)\end{aligned}$$

Similarly, for $\dot{C}_b$

$$\begin{aligned}\dot{C}_b &= -i\omega_b C_b+i\frac{|\wp_{ba}|\widetilde{E}}{\hbar}e^{i\phi}\cos(\nu t)C_a\\ &= -i\omega_b C_b+i\Omega_R e^{i\phi}\cos(\nu t)C_a. \quad (2.171)\end{aligned}$$

The solutions are obtained by expressing the equations of motion for the slowly varying amplitudes

$$c_a=C_a e^{i\omega_a t}, \quad (2.172)$$

$$c_b = C_b e^{i\omega_b t}, \tag{2.173}$$

From Eq. 2.170

$$\begin{aligned} \dot{C}_a &= -i\omega_a C_a + i\Omega_R e^{-i\phi}\cos(\nu t) C_b & (2.174) \\ \dot{C}_a e^{i\omega_a t} + i\omega_a C_a e^{i\omega_a t} &= i\Omega_R e^{-i\phi} C_b e^{i\omega_a t}\frac{1}{2}\left(e^{i\nu t} + e^{-i\nu t}\right) & (2.175) \\ \dot{\overline{C_a e^{i\omega_a t}}} &= i\frac{\Omega_R}{2} e^{-i\phi} c_b e^{i(\omega_a - \omega_b)t}\left(e^{i\nu t} + e^{-i\nu t}\right), & (2.176) \end{aligned}$$

and hence

$$\begin{aligned} \dot{c}_a &= i\frac{\Omega_R}{2} e^{-i\phi} c_b \left(e^{i(\omega+\nu)t} + e^{i(\omega-\nu)t}\right) & (2.177) \\ &= i\frac{\Omega_R}{2} e^{-i\phi} c_b e^{i(\omega-\nu)t} & (2.178) \\ &= i\frac{\Omega_R}{2} e^{-i\phi} c_b e^{i\Delta t}, & (2.179) \end{aligned}$$

where $\dot{\overline{C_a e^{i\omega_a t}}} = \dot{C}_a e^{i\omega_a t} + i\omega_a C_a e^{i\omega_a t}$ and the atomic transition frequency is defined as $\omega = \omega_a - \omega_b$. Detuning is $\Delta = \omega - \nu$. In the rotating wave approximation, counter rotating terms $e^{\pm i(\omega+\nu)t}$ are ignored because they are highly oscillating. Similar steps is performed on Eq. 2.171 getting

$$\begin{aligned} \dot{C}_b &= -i\omega_b C_b + i\Omega_R e^{i\phi}\cos(\nu t) C_a, & (2.180) \\ \dot{C}_b e^{i\omega_b t} + i\omega_b C_b e^{i\omega_b t} &= i\Omega_R e^{i\phi} c_a e^{-i\omega_a t} e^{i\omega_b t}\frac{1}{2}\left(e^{i\nu t} + e^{-i\nu t}\right), & (2.181) \end{aligned}$$

hence,

$$\begin{aligned} \dot{c}_b &= i\frac{\Omega_R}{2} e^{i\phi} c_a \left(e^{-i(\omega-\nu)t} + e^{-i(\omega+\nu)t}\right) & (2.182) \\ &= i\frac{\Omega_R}{2} e^{i\phi} c_a e^{-i\Delta t}. & (2.183) \end{aligned}$$

The solution for c_a and c_b are found using Laplace transform as below. Changes of variables are made.

$$\begin{aligned} \widetilde{c}_a &= c_a e^{-\frac{i\Delta t}{2}}, & (2.184) \\ \widetilde{c}_b &= c_b e^{\frac{i\Delta t}{2}}. & (2.185) \end{aligned}$$

From Eq. 2.179

$$\begin{aligned}
\dot{c}_a e^{\frac{-i\Delta t}{2}} &= i\frac{\Omega_R}{2}e^{-i\phi}c_b e^{\frac{i\Delta t}{2}} && (2.186)\\
c_a \dot{e}^{\frac{-i\Delta t}{2}} &= -\frac{i\Delta}{2}c_a e^{\frac{-i\Delta t}{2}} + i\frac{\Omega_R}{2}e^{-i\phi}c_b e^{\frac{i\Delta t}{2}} && (2.187)\\
\dot{\widetilde{c}}_a &= -\frac{i\Delta}{2}\widetilde{c}_a + i\frac{\Omega_R}{2}e^{-i\phi}\widetilde{c}_b, && (2.188)
\end{aligned}$$

using $c_a \dot{e}^{\frac{-i\Delta t}{2}} = \dot{c}_a e^{\frac{-i\Delta t}{2}} - \frac{i\Delta}{2}c_a e^{\frac{-i\Delta t}{2}}$. Similarly, we have

$$\begin{aligned}
\dot{c}_b e^{\frac{i\Delta t}{2}} &= i\frac{\Omega_R}{2}e^{i\phi}c_a e^{-\frac{i\Delta t}{2}} && (2.189)\\
c_b \dot{e}^{\frac{i\Delta t}{2}} &= \frac{i\Delta}{2}c_b e^{\frac{i\Delta t}{2}} + i\frac{\Omega_R}{2}e^{i\phi}c_a e^{-\frac{i\Delta t}{2}} && (2.190)\\
\dot{\widetilde{c}}_b &= \frac{i\Delta}{2}\widetilde{c}_b + i\frac{\Omega_R}{2}e^{i\phi}\widetilde{c}_a. && (2.191)
\end{aligned}$$

By performing Laplace transform, let use the identity $L\{f'(x)\} = sF(s) - f(0)$ and yield

$$\begin{aligned}
\dot{\widetilde{c}}_a &= -\frac{i\Delta}{2}\widetilde{c}_a + i\frac{\Omega_R}{2}e^{-i\phi}\widetilde{c}_b && (2.192)\\
s\widetilde{c}_a(s) - \widetilde{c}_a(0) &= -\frac{i\Delta}{2}\widetilde{c}_a(s) + i\frac{\Omega_R}{2}e^{-i\phi}\widetilde{c}_b(s) && (2.193)\\
\left[s + \frac{i\Delta}{2}\right]\widetilde{c}_a(s) &= \widetilde{c}_a(0) + i\frac{\Omega_R}{2}e^{-i\phi}\widetilde{c}_b(s). && (2.194)
\end{aligned}$$

Meanwhile, the expression for $\widetilde{c}_b(s)$ is obtained

$$\begin{aligned}
\dot{\widetilde{c}}_b &= \frac{i\Delta}{2}\widetilde{c}_b + i\frac{\Omega_R}{2}e^{i\phi}\widetilde{c}_a && (2.195)\\
s\widetilde{c}_b(s) - \widetilde{c}_b(0) &= \frac{i\Delta}{2}\widetilde{c}_b(s) + i\frac{\Omega_R}{2}e^{i\phi}\widetilde{c}_a(s) && (2.196)\\
\widetilde{c}_b(s) &= \frac{\widetilde{c}_b(0)}{\left(s - \frac{i\Delta}{2}\right)} + i\frac{\Omega_R}{2\left(s - \frac{i\Delta}{2}\right)}e^{i\phi}\widetilde{c}_a(s), && (2.197)
\end{aligned}$$

Then, we insert Eq. 2.197 to Eq. 2.194 and yield

$$\begin{aligned}
\left(s + \frac{i\Delta}{2} + \frac{\Omega_R^2}{4\left(s - \frac{i\Delta}{2}\right)}\right)\widetilde{c}_a(s) &= \widetilde{c}_a(0) + i\frac{\Omega_R}{2\left(s - \frac{i\Delta}{2}\right)}\widetilde{c}_b(0)e^{-i\phi} && (2.198)\\
\left(\frac{4s^2 + \Delta^2 + \Omega_R^2}{4s - 2i\Delta}\right)\widetilde{c}_a(s) &= \widetilde{c}_a(0) + i\frac{\Omega_R}{(2s - i\Delta)}\widetilde{c}_b(0)e^{-i\phi}, && (2.199)
\end{aligned}$$

and the expression for $\widetilde{c}_a(s)$ is

$$\widetilde{c}_a(s) = \widetilde{c}_a(0)\frac{\left(s-\frac{i\Delta}{2}\right)}{\left[s^2+\left(\frac{1}{2}\sqrt{\Delta^2+\Omega_R^2}\right)^2\right]} + i\frac{\Omega_R}{2\left[s^2+\left(\frac{1}{2}\sqrt{\Delta^2+\Omega_R^2}\right)^2\right]}\widetilde{c}_b(0)e^{-i\phi}. \quad (2.200)$$

Let us define a new relation to connect the detuning and the Rabi frequency as the following expression

$$\Omega = \sqrt{\Delta^2+\Omega_R^2}, \quad (2.201)$$

and hence we substitute Eq. 2.201 into Eq. 2.200

$$\begin{aligned}\widetilde{c}_a(s) &= \widetilde{c}_a(0)\frac{\left(s-\frac{i\Delta}{2}\right)}{s^2+\left(\frac{1}{2}\Omega\right)^2} + i\frac{\Omega_R}{2\left[s^2+\left(\frac{1}{2}\Omega\right)^2\right]}\widetilde{c}_b(0)e^{-i\phi} &(2.202)\\ &= \widetilde{c}_a(0)\left[\frac{s}{s^2+\left(\frac{1}{2}\Omega\right)^2} - \frac{(i\Delta/2)\frac{1}{2}\Omega}{\frac{1}{2}\Omega\left[s^2+\left(\frac{1}{2}\Omega\right)^2\right]}\right] &(2.203)\\ &\quad + i\frac{\Omega_R\left(\frac{1}{2}\Omega\right)}{\Omega\left[s^2+\left(\frac{1}{2}\Omega\right)^2\right]}\widetilde{c}_b(0)e^{-i\phi},\end{aligned}$$

with

$$\widetilde{c}_a(t) = \widetilde{c}_a(0)\left[\cos\frac{\Omega t}{2} - \frac{i\Delta}{\Omega}\sin\frac{\Omega t}{2}\right] + i\frac{\Omega_R}{\Omega}\widetilde{c}_b(0)e^{-i\phi}\sin\frac{\Omega t}{2}.$$

Since $c_a = \widetilde{c}_a e^{\frac{i\Delta t}{2}}$, then we expand it as

$$c_a = \left\{c_a(0)\left[\cos\frac{\Omega t}{2} - \frac{i\Delta}{\Omega}\sin\frac{\Omega t}{2}\right] + i\frac{\Omega_R}{\Omega}c_b(0)e^{-i\phi}\sin\frac{\Omega t}{2}\right\}e^{\frac{i\Delta t}{2}} \quad (2.204)$$

In order to find c_b, we perform the operation

$$\left[s-\frac{i\Delta}{2}+\frac{\Omega_R^2}{4\left(s+\frac{i\Delta}{2}\right)}\right]\widetilde{c}_b(s) = \widetilde{c}_b(0) + i\frac{\Omega_R}{2\left(s+\frac{i\Delta}{2}\right)}e^{i\phi}\widetilde{c}_a(0),$$

and yield

$$\widetilde{c}_b(s) = \frac{\left(s+\frac{i\Delta}{2}\right)}{s^2+\frac{1}{4}\left(\Delta^2+\Omega_R^2\right)}\widetilde{c}_b(0) + i\frac{\Omega_R}{2\left(s^2+\frac{1}{4}\left(\Delta^2+\Omega_R^2\right)\right)}e^{i\phi}\widetilde{c}_a(0) \quad (2.205)$$

Again, we substitute Eq. 2.201 into Eq. 2.205

$$\widetilde{c}_b(s) = \frac{\left(s+\frac{i\Delta}{2}\right)}{s^2+\left(\frac{1}{2}\Omega\right)^2}\widetilde{c}_b(0)+i\frac{\Omega_R}{2\left(s^2+\left(\frac{1}{2}\Omega\right)^2\right)}e^{i\phi}\widetilde{c}_a(0) \tag{2.206}$$

$$= \widetilde{c}_b(0)\left[\frac{s}{s^2+\left(\frac{1}{2}\Omega\right)^2}+\frac{i\Delta}{\Omega}\frac{\frac{1}{2}\Omega}{\left(s^2+\left(\frac{1}{2}\Omega\right)^2\right)}\right] + i\frac{\Omega_R}{\Omega}\frac{\frac{1}{2}\Omega}{\left(s^2+\left(\frac{1}{2}\Omega\right)^2\right)}e^{i\phi}\widetilde{c}_a(0) \tag{2.207}$$

$$= \widetilde{c}_b(0)\left[\cos\frac{\Omega t}{2}+\frac{i\Delta}{\Omega}\sin\frac{\Omega t}{2}\right]+i\frac{\Omega_R}{\Omega}e^{i\phi}\widetilde{c}_a(0)\sin\frac{\Omega t}{2} \tag{2.208}$$

Since $c_b=\widetilde{c}_b e^{-\frac{i\Delta t}{2}}$, then

$$c_b=\left\{c_b(0)\left[\cos\frac{\Omega t}{2}+\frac{i\Delta}{\Omega}\sin\frac{\Omega t}{2}\right]+i\frac{\Omega_R}{\Omega}e^{i\phi}c_a(0)\sin\frac{\Omega t}{2}\right\}e^{-\frac{i\Delta t}{2}} \tag{2.209}$$

- Note that conservation of probability $|c_a(t)|^2+|c_b(t)|^2=1$ since the atom is in state $|a\rangle$ or $|b\rangle$. If the atom is assumed to be in state $|a\rangle$ initially, $c_a(0)=1$ and $c_b(0)=0$. The probabilities of the atom being in states $|a\rangle$ and $|b\rangle$ at time t are defined by $|c_a(t)|^2$ and $|c_b(t)|^2$ respectively. The inversion is

$$W(t) = |c_a(t)|^2-|c_b(t)|^2 \tag{2.210}$$

$$= \left|\left[\cos\frac{\Omega t}{2}-\frac{i\Delta}{\Omega}\sin\frac{\Omega t}{2}\right]e^{\frac{i\Delta t}{2}}\right|^2-\left|i\frac{\Omega_R}{\Omega}e^{i\phi}\sin\frac{\Omega t}{2}e^{-\frac{i\Delta t}{2}}\right|^2 \tag{2.211}$$

$$= \left(\frac{\Delta^2-\Omega_R^2}{\Omega^2}\right)\sin^2\left(\frac{\Omega t}{2}\right)+\cos^2\left(\frac{\Omega t}{2}\right) \tag{2.212}$$

A dipole moment is induced between the two atomic levels as a response to interaction towards incident field and it is depicted by the expectation value of dipole moment operator

$$P(t) = e\langle\psi(t)|r|\psi(t)\rangle \tag{2.213}$$

$$= e\left[\langle a|C_a^*(t)+\langle b|C_b^*(t)\right]r\left[C_a(t)|a\rangle+C_b(t)|b\rangle\right] \tag{2.214}$$

$$= e\langle a|r|b\rangle C_a^*(t)C_b(t)+e\langle b|r|a\rangle C_a(t)C_b^*(t) \tag{2.215}$$

$$= C_a^*C_b\wp_{ab}+c.c.=c_a^*c_b\wp_{ab}e^{i\omega t}+c.c. \tag{2.216}$$

We use $|\psi(t)\rangle=C_a(t)|a\rangle+C_b(t)|b\rangle$ and $\wp_{ab}=e\langle a|x|b\rangle=\wp_{ba}^*$. For an atom initially in the upper level

$$
\begin{aligned}
P(t) &= \left\{\left[\cos\frac{\Omega t}{2}+\frac{i\Delta}{\Omega}\sin\frac{\Omega t}{2}\right]\right\} e^{\frac{-i\Delta t}{2}} \left\{ i\frac{\Omega_R}{\Omega}e^{i\phi}\sin\frac{\Omega t}{2}\right\} e^{-\frac{i\Delta t}{2}} \wp_{ab} e^{i\omega t} + c.c. \\
&= i\frac{\Omega_R}{\Omega}\wp_{ab}\left[\cos\frac{\Omega t}{2}+\frac{i\Delta}{\Omega}\sin\frac{\Omega t}{2}\right]\sin\frac{\Omega t}{2}e^{i\phi}e^{i\nu t} + c.c. \\
&= 2Re\left\{\frac{i\Omega_R}{\Omega}\wp_{ab}\left[\cos\left(\frac{\Omega t}{2}\right)+\frac{i\Delta}{\Omega}\sin\left(\frac{\Omega t}{2}\right)\right]\sin\left(\frac{\Omega t}{2}\right)e^{i\phi}e^{i\nu t}\right\} \qquad (2.217)
\end{aligned}
$$

The dipole moment therefore oscillates with the frequency of the incident field. For case where atom is at resonance with the incident field $\Delta = 0$, causing $\Omega = \Omega_R$. Hence

$$
\begin{aligned}
W(t) &= -\sin^2\left(\frac{\Omega t}{2}\right)+\cos^2\left(\frac{\Omega t}{2}\right) \\
&= 2\cos^2\left(\frac{\Omega t}{2}\right)-1 \\
&= \cos\left(\Omega_R t\right) \qquad (2.218)
\end{aligned}
$$

which indicates that the inversion oscillates between -1 and 1 at frequency Ω_R. The atom experiences Rabi (Brune et al., 1996) flopping between the upper and lower levels due to interaction with electromagnetic field.

2. Unitary Transformation in Interaction Picture The Scrodinger equation is expressed in the form of

$$
\frac{\partial}{\partial t}\left|\psi(t)\right\rangle = -\frac{i}{\hbar}H\left|\psi(t)\right\rangle, \qquad (2.219)
$$

where it can be integrated to give

$$
\begin{aligned}
\left|\psi(t)\right\rangle &= -\frac{i}{\hbar}\int HU(t)\left|\psi(t)\right\rangle \partial t \qquad (2.220) \\
&= U(t)\left|\psi(0)\right\rangle, \qquad (2.221)
\end{aligned}
$$

as the unitary time-evolution operator (Yuen, 1976) is defined to be

$$
\dot{U}(t) = -\frac{i}{\hbar}HU(t), \qquad (2.222)
$$

where $U(0) = 1$. In the interaction picture, the time dependence is assigned to the state vector due to the interaction energy. The state vector $\left|\psi_I\right\rangle$ in the interaction picture

$$
\left|\psi_I(t)\right\rangle = U_0^{+}(t)\left|\psi(t)\right\rangle, \qquad (2.223)
$$

with

$$
U_0(t) = \exp\left(-\frac{i}{\hbar}H_0 t\right). \qquad (2.224)
$$

Thus, we have

$$\frac{\partial}{\partial t}\left|\psi_I(t)\right\rangle = \frac{\partial}{\partial t}\left[U_0^+(t)\left|\psi(t)\right\rangle\right] \tag{2.225}$$

$$= \left[\frac{\partial}{\partial t}U_0^+(t)\right]\left|\psi(t)\right\rangle + U_0^+(t)\frac{\partial}{\partial t}\left|\psi(t)\right\rangle, \tag{2.226}$$

and hence

$$\frac{\partial}{\partial t}\left|\psi_I(t)\right\rangle = \left[\frac{i}{\hbar}H_0\exp\left(\frac{i}{\hbar}H_0 t\right)\right]\left|\psi(t)\right\rangle - \frac{i}{\hbar}HU_0^+(t)\left|\psi(t)\right\rangle \tag{2.227}$$

$$= \frac{i}{\hbar}H_0U_0^+(t)\left|\psi(t)\right\rangle - \frac{i}{\hbar}H\left|\psi_I(t)\right\rangle \tag{2.228}$$

$$= \frac{i}{\hbar}\left|\psi_I(t)\right\rangle(H_0 - H) \tag{2.229}$$

$$= -\frac{i}{\hbar}U_0^+(t)H_1U_0(t)\left|\psi_I(t)\right\rangle. \tag{2.230}$$

In other word, we can express the derivative in this form

$$\frac{\partial}{\partial t}\left|\psi_I(t)\right\rangle = -\frac{i}{\hbar}V(t)\left|\psi_I(t)\right\rangle \tag{2.231}$$

and the interaction picture Hamiltonian is defined as

$$V(t) = U_0^+(t)H_1U_0(t), \tag{2.232}$$

The transformation of an operator O in the Scrodinger picture follows as $O_I(t) = U_0^+(t)OU_0(t)$. The expectation value is

$$\langle O\rangle = \langle\psi(t)|O|\psi(t)\rangle \tag{2.233}$$

$$= \left\langle\psi(t)|U_0^+(t)OU_0(t)|\psi(t)\right\rangle \tag{2.234}$$

$$= \langle\psi(t)|O_I|\psi(t)\rangle. \tag{2.235}$$

The solution for Eq. 2.231

$$\left|\psi_I(t)\right\rangle = \left|\psi_I(0)\right\rangle\Gamma\exp\left(-\frac{i}{\hbar}\int_0^t V(\tau)\,\partial\tau\right) \tag{2.236}$$

$$= U_I(t)\left|\psi_I(0)\right\rangle, \tag{2.237}$$

where the time-evolution operator in the interaction picture is $U_I(t) = \Gamma\exp\left[-\frac{i}{\hbar}\int_0^t V(\tau)\,d\tau\right]$ and Γ is the time ordering operator. $\Gamma\exp\left(-\frac{i}{\hbar}\int_0^t V(\tau)\,\partial\tau\right)$ is a shorthand notation of $\Gamma\exp\left[-\frac{i}{\hbar}\int_0^t V(\tau)\,d\tau\right] = 1 - \frac{i}{\hbar}\int_0^t dt_1 V(t_1) + \left(-\frac{i}{\hbar}\right)^2\int_0^t dt_1\int_0^{t_1} dt_2 V(t_1)V(t_2) + ...$ Let us consider interaction between two level atom and a monochromatic field with frequency v, where the Hamiltonian is described by $H = H_0 + H_1$

$$H_0 = \hbar\omega_a |a\rangle\langle a| + \hbar\omega_b |b\rangle\langle b| \tag{2.238}$$

$$H_0^n = (\hbar\omega_a)^n |a\rangle\langle a| + (\hbar\omega_b)^n |b\rangle\langle b| \tag{2.239}$$

$$H_1 = -\left(\wp_{ab}|a\rangle\langle b| + \wp_{ba}|b\rangle\langle a|\right)E(t), \tag{2.240}$$

Hence, the transformation operator becomes

$$U_0(t) = \exp\left(-\frac{i}{\hbar}H_0 t\right) \tag{2.241}$$

$$= \exp\left(-\frac{i}{\hbar}\left[\hbar\omega_a|a\rangle\langle a| + \hbar\omega_b|b\rangle\langle b|\right]t\right) \tag{2.242}$$

$$= \exp(-i\omega_a t)|a\rangle\langle a| + \exp(-i\omega_b t)|b\rangle\langle b|. \tag{2.243}$$

The interaction picture Hamiltonian, $V(t)$ for an atom at $z = 0$ is

$$V(t) = U_0^+(t) H_1 U_0(t) \tag{2.244}$$

$$= U_0^+(t)\left[-\left(\wp_{ab}|a\rangle\langle b| + \wp_{ba}|b\rangle\langle a|\right)E(t)\right]U_0(t) \tag{2.245}$$

$$= U_0^+(t)\left[-\wp_{ab}e^{i\phi}e^{-i\phi}|a\rangle\langle b|\widetilde{E} - \wp_{ba}e^{-i\phi}e^{i\phi}|b\rangle\langle a|\widetilde{E}\right] \tag{2.246}$$
$$\times\cos(vt)U_0(t),$$

and we further expand Eq. 2.246 and yield

$$V(t) = U_0^+(t)\left[-\hbar\frac{|\wp_{ab}|\widetilde{E}}{\hbar}e^{-i\phi}|a\rangle\langle b| - \hbar\frac{|\wp_{ba}|\widetilde{E}}{\hbar}e^{i\phi}|b\rangle\langle a|\right] \tag{2.247}$$
$$\times\cos(vt)U_0(t)$$

$$= -\hbar\Omega_R U_0^+(t)\left[e^{-i\phi}|a\rangle\langle b| + e^{i\phi}|b\rangle\langle a|\right]U_0(t)\cos(vt). \tag{2.248}$$

Then, the unitary operator as in Eq. 2.243 is plucked into Eq. 2.248

$$V(t) = -\frac{\hbar\Omega_R}{2}\left(e^{i\omega_a t}|a\rangle\langle a| + e^{i\omega_b t}|b\rangle\langle b|\right)\left[e^{-i\phi}|a\rangle\langle b| + e^{i\phi}|b\rangle\langle a|\right] \tag{2.249}$$
$$\times\left[e^{-i\omega_a t}|a\rangle\langle a| + e^{-i\omega_b t}|b\rangle\langle b|\right]\left(e^{ivt} + e^{-ivt}\right)$$

$$= -\frac{\hbar\Omega_R}{2}\{(e^{i(\omega+v)t}|a\rangle\langle a||b\rangle\langle b| + e^{-i\Delta t}|b\rangle\langle b||a\rangle\langle a| + e^{i\Delta t}|a\rangle\langle a||b\rangle\langle b|$$
$$+e^{-i(\omega+v)t}|b\rangle\langle b||a\rangle\langle a|) \times \left[e^{-i\phi}|a\rangle\langle b| + e^{i\phi}|b\rangle\langle a|\right]\}, \tag{2.250}$$

by expanding Eq. 2.250

$$\begin{aligned}V(t) &= -\frac{\hbar\Omega_R}{2}\{e^{i(\omega+v)t}e^{-i\phi}|a\rangle\langle a||a\rangle\langle b||b\rangle\langle b|+e^{-i\Delta t}e^{-i\phi}|b\rangle\langle b||a\rangle\langle b||a\rangle\langle a| \\ &+e^{i\Delta t}e^{-i\phi}|a\rangle\langle a||a\rangle\langle b||b\rangle\langle b|+e^{-i(\omega+v)t}e^{-i\phi}|b\rangle\langle b||a\rangle\langle b||a\rangle\langle a| \\ &+e^{i(\omega+v)t}e^{i\phi}|a\rangle\langle a||b\rangle\langle a||b\rangle\langle b|+e^{-i\Delta t}e^{i\phi}|b\rangle\langle b||b\rangle\langle a||a\rangle\langle a| \\ &+e^{i\Delta t}e^{i\phi}|a\rangle\langle a||b\rangle\langle a||b\rangle\langle b|+e^{-i(\omega+v)t}e^{i\phi}|b\rangle\langle b||b\rangle\langle a||a\rangle\langle a|\}. \qquad (2.251)\end{aligned}$$

Finally, after simplify the above expression, we have

$$V(t) = -\frac{\hbar\Omega_R}{2}\left\{e^{i(\omega+v)t}e^{-i\phi}|a\rangle\langle b|+e^{i\Delta t}e^{-i\phi}|a\rangle\langle b|+e^{-i\Delta t}e^{i\phi}|b\rangle\langle a|+e^{-i(\omega+v)t}e^{i\phi}|b\rangle\langle a|\right\}, \qquad (2.252)$$

where $E(t) = \widetilde{E}\cos(vt)\,\wp_{ba} = |\wp_{ba}|\exp(i\phi)$, $\wp_{ab} = |\wp_{ab}|\exp(-i\phi)$, Rabi frequency is $\Omega_R = \frac{|\wp_{ba}|E}{\hbar}$, $\omega = \omega_a - \omega_b$, and $\Delta = \omega - v$. The term proportional to $\exp(i(\omega+v)t)$ vary very rapidly can be dropped in the rotating wave approximation. Therefore, with resonance $\Delta = 0$,

$$V(t) = -\frac{\hbar\Omega_R}{2}\left(e^{-i\phi}|a\rangle\langle b|+e^{i\phi}|b\rangle\langle a|\right). \qquad (2.253)$$

3. Rotating-Wave Approximation Rotating wave approximation (Zaheer & Zubairy, 1988) is used to keep only energy conserving terms in the Hamiltonian. The counter-rotating terms are always dropped out because they never show up om exact situations. Consider atom is placed at the origin such that $\mathbf{R} = 0$, the interaction picture Hamiltonian can be obtained using dipole approximation

$$H_1 = -e\mathbf{r}.\mathbf{E}(\mathbf{r}_0,t), \qquad (2.254)$$

then we can perform the unitary transformation on Eq. 2.254

$$\begin{aligned}V(t) &= U_0^+(t)H_1U_0(t) &(2.255)\\ &= -ee^{\frac{i}{\hbar}H_0t}\mathbf{r}e^{-\frac{i}{\hbar}H_0t}\cdot\mathbf{E}(t) &(2.256)\\ &= -e\mathbf{r}(t)\cdot\mathbf{E}(t) &(2.257)\end{aligned}$$

where the unitary operator is defined as

$$U_0(t) = e^{-\frac{i}{\hbar}H_0 t}, \tag{2.258}$$

and the vector $\mathbf{r}$ is

$$\mathbf{r}(t) = e^{\frac{i}{\hbar}H_0 t}\mathbf{r}e^{-\frac{i}{\hbar}H_0 t}. \tag{2.259}$$

Hence, two equations can be obtained, which is

$$\begin{aligned} V_{ab}(t) &= -e\mathbf{r}_{ab}(t).\mathbf{E}(t) && (2.260) \\ &= -e\mathbf{r}_{ab}.\mathbf{E}(t)e^{i\omega t}, && (2.261) \end{aligned}$$

and

$$\begin{aligned} V_{ba}(t) &= -e\mathbf{r}_{ba}(t).\mathbf{E}(t) && (2.262) \\ &= -e\mathbf{r}_{ba}.\mathbf{E}(t)e^{-i\omega t}, && (2.263) \end{aligned}$$

where ω is the atomic frequency. Consider for the case of linear polarization where $\mathbf{E}(t) = \widehat{x}\widetilde{E}\cos\nu t$, then Eq. 2.261 becomes

$$\begin{aligned} V_{ab}(t) &= -e\mathbf{r}_{ab}.\widehat{x}\widetilde{E}\cos\nu t e^{i\omega t} && (2.264) \\ &= -ex_{ab}\frac{\widetilde{E}}{2}\left(e^{i(\nu+\omega)t} + e^{-i(\nu-\omega)t}\right) && (2.265) \\ &\simeq -ex_{ab}\frac{\widetilde{E}}{2}e^{-i(\nu-\omega)t}. && (2.266) \end{aligned}$$

Similarly, Eq. 2.263 becomes,

$$\begin{aligned} V_{ba}(t) &= -e\mathbf{r}_{ba}.\widehat{x}\widetilde{E}\cos\nu t e^{-i\omega t} && (2.267) \\ &= -ex_{ba}\frac{\widetilde{E}}{2}\left(e^{i(\nu-\omega)t} + e^{-i(\nu+\omega)t}\right) && (2.268) \\ &\simeq -ex_{ba}\frac{\widetilde{E}}{2}e^{i(\nu-\omega)t}. && (2.269) \end{aligned}$$

The rotating wave approximation is established by neglecting the counter rotating terms which is fast rotating, $\exp[\pm i(\nu+\omega)t]$. For the case of left-circular polarization, the electric field is given by $\mathbf{E}(t) = \widehat{x}\widetilde{E}\cos\nu t - \widehat{y}\widetilde{E}\sin\nu t$. Thus

$$\begin{aligned} V_{ab}(t) &= \left[-e\mathbf{r}_{ab}.\widehat{x}\widetilde{E}\cos\nu t + e\mathbf{r}_{ab}.\widehat{y}\widetilde{E}\sin\nu t\right]e^{i\omega t} && (2.270) \\ &= -e\widetilde{E}\left(x_{ab}\cos\nu t - y_{ab}\sin\nu t\right)e^{i\omega t}, && (2.271) \end{aligned}$$

and

$$
\begin{aligned}
V_{ba}(t) &= \left[-e\mathbf{r}_{ba}.\widehat{x}\widetilde{E}\cos\nu t + e\mathbf{r}_{ba}.\widehat{y}\widetilde{E}\sin\nu t\right]e^{-i\omega t} && (2.272)\\
&= -e\widetilde{E}\left(x_{ba}\cos\nu t - y_{ba}\sin\nu t\right)e^{-i\omega t}. && (2.273)
\end{aligned}
$$

As $ex_{ba} = \wp$ and $ey_{ba} = i\wp$, we get the final expression for $V_{ab}(t)$

$$
\begin{aligned}
V_{ab}(t) &= -\wp\widetilde{E}\left(\cos\nu t - i\sin\nu t\right)e^{i\omega t} && (2.274)\\
&= -\wp\widetilde{E}e^{-i(\nu-\omega)t} && (2.275)
\end{aligned}
$$

and in the meantime, the final expression for $V_{ba}(t)$ is

$$
\begin{aligned}
V_{ba}(t) &= -\wp\widetilde{E}\left(\cos\nu t + i\sin\nu t\right)e^{-i\omega t} && (2.276)\\
&= -\wp\widetilde{E}e^{i(\nu-\omega)t} && (2.277)
\end{aligned}
$$

As can be seen, the counter rotating terms does not appear in both expression.

CHAPTER 3

HIGH FIELD PROCESSES

In this chapter, a brief description on the development that leads to the discovery of the natural phenomenon which occurred during the interaction between intense light fields and matter. These basic phenomenon include above threshold ionization (ATI), multi-photon ionization (MPI) and high harmonic generation (HHG) (Sheehy et al., 1999). The theory of each phenomenon will be discussed in the following sections. In the previous chapter, we have discussed the perturbation theory (Simon, 1973) on transition amplitudes. Nevertheless, in this chapter the limit of perturbation theory will be shown and as a result, a new description which is known as non-perturbative theory is needed in order to interpret the phenomenon by intense laser field (Federov, 1991). In the following section we will consider the main ideas of the conventional perturbation theory and its failure when the field becomes very strong due to the appearance of the relativistic effect. We will discuss this in chapter 5 on generalizing the perturbative photionization model by taking consideration of the relativistic effect.

In spite of that, the nonperturbative model for the intense-field processes (Mittleman, 1993) and point out several important nonlinear parameters that emerge naturally from it and the significance of the so-called "single active electron" (SAE) hypothesis in single-electron processes in intense fields.

3.1 Above Threshold Ionization

In quantum mechanics ionization of the atom with the electromagnetic radiation, with violation of Einstein formula, i.e. when kinetic energy of the emitted electrons is larger than the difference between the photon energy and the ionization energy or the work function. In that case the generalized Einstein formula is valid

$$n\hbar\omega = W + E_k \tag{3.1}$$

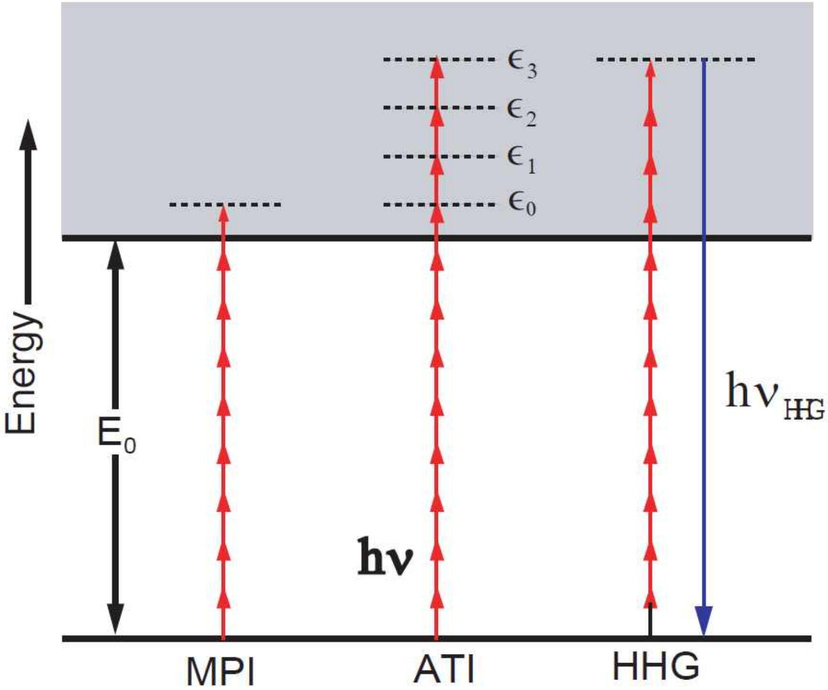


Figure 3.1: Several processes occured in the Strong Field Ionization

where n is the arbitrary integer number, W is the ionization energy (work function) and E_k is the electron kinetic energy.

This phenomenon is measurable if and only if the electromagnetic field is comparable with the field which keeps the electrons in the atom, for example generated with a very strong laser.

In principal, ATI (Corkum et al., 1989) is a process in which atoms absorb more than the minimum number of photons required to be ionized. The ATI (Freeman et al., 1987) spectrum consists of a series of peaks equally separated by the photon energy. Hence ATI may be explained by solving the time-dependent Schrödinger equation in the approximate manner. The Schrödinger equation for the free electron in the field of the electromagnetic wave in one dimension and in the radiation gauge is given by

$$i\hbar\frac{\partial\Psi}{\partial t} = \frac{1}{2m}\left[\frac{\hbar}{i}\nabla - eA\left(t\right)\right]^2\Psi, \tag{3.2}$$

where

$$A\left(t\right) = \frac{E_0}{\omega}\cos\left(\omega t\right). \tag{3.3}$$

and the electric field is given by

$$E(t) = -\frac{\partial A}{\partial t} = E_0 \sin(\omega t) \tag{3.4}$$

We make an approximation that the wavefunction $\Psi = e^{-C(t)+ikx}$, then we substitute into the Schrödinger equation and yield

$$i\hbar \frac{\partial}{\partial t} e^{iC(t)+ikx} = \frac{1}{2m}\left[\frac{\hbar}{i}\nabla - eA(t)\right]^2 e^{iC(t)+ikx}. \tag{3.5}$$

In one-dimensional case, we have

$$i\dot{C}(t)\, i\hbar e^{iC(t)+ikx} = \frac{1}{2m}\left[-\hbar^2\nabla^2 - \frac{\hbar}{i}e\nabla\cdot A(t) - \frac{\hbar}{i}eA(t)\nabla + e^2A(t)^2\right] e^{iC(t)+ikx} \tag{3.6}$$

$$= \frac{1}{2m}\left[-\hbar^2 (ik)^2 - \frac{\hbar}{i}e\nabla\cdot A(t) - \frac{\hbar}{i}eA(t)\, ik - \frac{\hbar}{i}eA(t)\, ik + e^2A(t)^2\right] \times e^{iC(t)+ikx}. \tag{3.7}$$

However, in Coulomb gauge where $\nabla \cdot A(t) = 0$, equation 3.6 becomes,

$$-\dot{C}(t)\,\hbar e^{iC(t)+ikx} = \frac{1}{2m}\left[\hbar^2k^2 - 2\frac{\hbar}{i}eA(t)\, ik + e^2A(t)^2\right] e^{iC(t)+ikx}, \tag{3.8}$$

$$-\dot{C}(t)\,\hbar = \frac{1}{2m}\left[\hbar^2k^2 - 2\hbar eA(t)\, k + e^2A(t)^2\right]. \tag{3.9}$$

After expanding, we obtain

$$-\dot{C}(t)\,\hbar = \frac{1}{2m}\left[\hbar^2k^2 - eA(t)\right]^2. \tag{3.10}$$

3.2 Tunneling Ionization

In order to understand the tunneling ionization (Fittinghoff, Bolton, Chang, & Kulander, 1994) process, firstly, the length-gauge Hamiltonian (Fabrikant & Gallup, 2009) for such system is

$$H_{LG} = \frac{\mathbf{p}^2}{2m} + e\mathbf{F}\cdot\mathbf{z}, \tag{3.11}$$

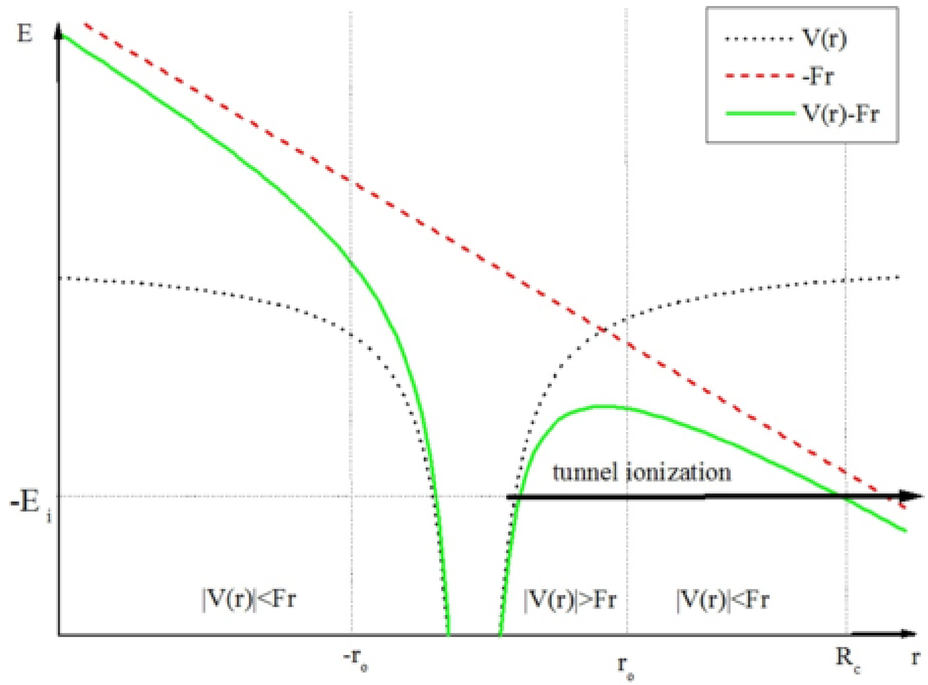


Figure 3.2: The process of tunneling ionization

where **p** is the kinetic momentum, **F** is the electric field strength and **z** is the direction of the field.

The boundary condition as below states that at some point z_0 the WKB wave function(Muth-Böhm, Becker, & Faisal, 2000) must match with the wave function of the bound system

$$\Psi(z_0) = \Psi_0(z_0) . \tag{3.12}$$

The mixed representation wave function (WKB wave function near a caustic)

$$\Psi(x,y,z) = \frac{1}{2\pi}\int dp_x \int dp_y \Phi(p_x,p_y,p_z) \exp\left(\frac{ixp_x + iyp_y}{\hbar}\right), \tag{3.13}$$

where

$$\Phi(p_x,p_y,p_z) = \frac{1}{2\pi}\int dx \int dy \Psi(x,y,z) \exp\left(\frac{-ixp_x - iyp_y}{\hbar}\right). \tag{3.14}$$

We insert the Eq. 3.124 into Schrödinger eqn and yield

$$H_{LG}\Psi(x,y,z) = -I_p\Psi(x,y,z) . \tag{3.15}$$

where I_p is the ionization potential (the binding energy of the bound state).

For a short-range potential, the 3-dimentional equation is reduced to a single dimension (z-direction)

$$\begin{aligned}\Phi(p_x,p_y,z) &= \frac{1}{2\pi}\int dx\int dy \exp\left(\frac{-ixp_x-iyp_y}{\hbar}\right)\Psi(z)\\ &= \frac{1}{2\pi}\Psi(z)\frac{i\hbar}{p_x}\frac{i\hbar}{p_y}\exp\left(\frac{-ixp_x-iyp_y}{\hbar}\right)\\ &= -\frac{1}{2\pi}\frac{\hbar^2}{p_xp_y}\Psi(z)\exp\left(\frac{-ixp_x-iyp_y}{\hbar}\right), \qquad (3.16)\end{aligned}$$

where the wavefunction is defined as

$$\Psi(z) = -\frac{2\pi p_xp_y}{\hbar^2}\Phi(p_x,p_y,z)\exp\left(\frac{ixp_x+iyp_y}{\hbar}\right). \qquad (3.17)$$

Then, the Hamiltonian of the system is

$$\begin{aligned}H_{LG}\Psi(z) &= \left[-\frac{\hbar^2}{2m}\nabla^2+e\mathbf{F}\cdot\mathbf{z}\right]\Psi(z) \qquad (3.18)\\ &= \frac{\hbar^2}{2m}\nabla^2\left[\frac{2\pi p_xp_y}{\hbar^2}\Phi(p_x,p_y,z)\exp\left(\frac{ixp_x+iyp_y}{\hbar}\right)\right] \qquad (3.19)\\ &\quad -e\mathbf{F}\cdot\mathbf{z}\left[\frac{2\pi p_xp_y}{\hbar^2}\Phi(p_x,p_y,z)\exp\left(\frac{ixp_x+iyp_y}{\hbar}\right)\right].\end{aligned}$$

After expanding, Eq. 3.18 becomes,

$$\begin{aligned}H_{LG}\Psi(z) &= \left[-\frac{p_x^2}{2m}-\frac{p_y^2}{2m}\right]\frac{2\pi p_xp_y}{\hbar^2}\exp\left(\frac{ixp_x+iyp_y}{\hbar}\right)\Phi(p_x,p_y,z) \qquad (3.20)\\ &\quad +\frac{1}{2m}2\pi p_xp_y\exp\left(\frac{ixp_x+iyp_y}{\hbar}\right)\frac{\partial^2}{\partial z^2}\Phi(p_x,p_y,z)\\ &\quad -e\mathbf{F}\cdot\mathbf{z}\frac{2\pi p_xp_y}{\hbar^2}\exp\left(\frac{ixp_x+iyp_y}{\hbar}\right)\Phi(p_x,p_y,z).\end{aligned}$$

Since that

$$\begin{aligned}H_{LG}\Psi(x,y,z) &= -I_p\Psi(x,y,z)\\ &= I_p\frac{2\pi p_xp_y}{\hbar^2}\exp\left(\frac{ixp_x+iyp_y}{\hbar}\right)\Phi(p_x,p_y,z), \qquad (3.21)\end{aligned}$$

imply that

$$I_p\Phi(p_x,p_y,z) = \left[-\frac{p_x^2}{2m}-\frac{p_y^2}{2m}\right]\Phi(p_x,p_y,z) + \frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2}\Phi(p_x,p_y,z) - e\mathbf{F}\cdot\mathbf{z}\Phi(p_x,p_y,z) \quad (3.22)$$

and

$$\begin{aligned}\frac{\hbar^2}{2m}\frac{\partial^2\Phi(p_x,p_y,z)}{\partial z^2} &= \left[\frac{p_x^2}{2m}+\frac{p_y^2}{2m}\right]\Phi(p_x,p_y,z)+I_p\Phi(p_x,p_y,z)+e\mathbf{F}\cdot\mathbf{z}\Phi(p_x,p_y,z)\\ &= \left[\frac{p_x^2}{2m}+\frac{p_y^2}{2m}+I_p+e\mathbf{F}\cdot\mathbf{z}\right]\Phi(p_x,p_y,z), \quad (3.23)\end{aligned}$$

with the second order derivative take the form of

$$\begin{aligned}\frac{\partial^2\Phi(p_x,p_y,z)}{\partial z^2} &= \frac{2m}{\hbar^2}\left[\frac{p_x^2}{2m}+\frac{p_y^2}{2m}+I_p+e\mathbf{F}\cdot\mathbf{z}\right]\Phi(p_x,p_y,z) \quad (3.24)\\ &= \frac{2m}{\hbar^2}\left[E'+e\mathbf{F}\cdot\mathbf{z}\right]\Phi(p_x,p_y,z),\end{aligned}$$

where the expression

$$E' = \frac{p_x^2}{2m}+\frac{p_y^2}{2m}+I_p. \quad (3.25)$$

Then, we solve equation 3.24 and yield

$$\frac{\partial^2\Phi(p_x,p_y,z)}{\partial z^2} = \frac{2m}{\hbar^2}\left[E'+e\mathbf{F}\cdot\mathbf{z}\right]\Phi(p_x,p_y,z), \quad (3.26)$$

where

$$\Phi(p_x,p_y,z) = \frac{C}{\sqrt{p_z(z)}}e^{iS(p_x,p_y,z)/\hbar}, \quad (3.27)$$

with $S(p_x,p_y,z)$ is the classical action and $p_z(z) = \left|\partial S(p_x,p_y,z)/\partial z\right|$ is the kinetic momentum in the z-direction.

By using the WKB approximation, we imagine a particle of energy E moving through a region where the potential $V(x)$ is constant. If $E > V$, the wave function is of the form:

$$\begin{aligned}\psi(x) &= Ae^{\pm ikx}, \quad (3.28)\\ k &= \sqrt{\frac{2m(E-V)}{\hbar}}. \quad (3.29)\end{aligned}$$

We must take note that the essential idea of this method is suppose that $V(x)$ is not constant, but varies rather slowly in comparison to λ, so that the region containing many full wavelengths and the potential is essentially constant. Hence it is reasonable to suppose that ψ remains practically sinusoidal, except that the wavelength and the amplitude change slowly with x.

If $E < V$ and V is constant, then ψ is exponential

$$\psi(x) = Ae^{\pm\kappa x}, \tag{3.30}$$

$$\kappa = \sqrt{\frac{2m(V-E)}{\hbar}}. \tag{3.31}$$

Meanwhile, if $V(x)$ is not a constant but varies slowly in comparison with $1/\kappa$, then the solution remains practically exponential, except that A and κ are now slowly-varying functions of x. Besides, there exists two classical turning point, where $E \approx V$. For here, λ or $1/\kappa$ goes infinity, and $V(x)$ can hardly be said to vary "slowly" in comparison.

In the classical region, the Schrödinger equation is defined as

$$\left[-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial x^2} + V(x)\right]\psi = E\psi, \tag{3.32}$$

and the second order derivative is known as

$$\frac{\partial^2\psi}{\partial x^2} = -\frac{p^2}{\hbar^2}\psi, \tag{3.33}$$

where

$$p(x) = \sqrt{2m[E - V(x)]}. \tag{3.34}$$

For this equation, we always take $E > V(x)$, so that $p(x)$ is always real and we call "this" as classical region where classically the particle is confined to this range of x. In general, ψ is some complex function so that we can retain the sinusoidal form

$$\psi(x) = A(x)e^{i\phi(x)}, \tag{3.35}$$

where both $A(x)$ and $\phi(x)$ are real.

Notation: using a prime to denote the derivative with respect of x

$$
\begin{aligned}
\frac{d\psi}{dx} &= A'e^{i\phi(x)} + i\phi' A e^{i\phi(x)} && (3.36)\\
\frac{d^2\psi}{dx^2} &= A''e^{i\phi(x)} + iA'\phi' e^{i\phi(x)} + i\left(\phi'' A + \phi' A'\right)e^{i\phi(x)} - \left(\phi'\right)^2 A e^{i\phi(x)} && (3.37)\\
&= \left[A'' + 2iA'\phi' + iA\phi'' - A\left(\phi'\right)^2\right]e^{i\phi(x)}.
\end{aligned}
$$

Hence we make comparison with Eq. 4.107, imply that

$$
\begin{aligned}
\left[A'' + 2iA'\phi' + iA\phi'' - A\left(\phi'\right)^2\right]e^{i\phi(x)} &= -\frac{p^2}{\hbar^2}A(x)\,e^{i\phi(x)}, && (3.38)\\
A'' - A\left(\phi'\right)^2 + i\left[2A'\phi' + A\phi''\right] &= -\frac{p^2}{\hbar^2}A. && (3.39)
\end{aligned}
$$

This can be separated into two equations, one for real part

$$
\begin{aligned}
A'' - A\left(\phi'\right)^2 &= -\frac{p^2}{\hbar^2}A,\\
A'' &= A\left[\left(\phi'\right)^2 - \frac{p^2}{\hbar^2}\right], && (3.40)
\end{aligned}
$$

and one for imaginary part

$$
\begin{aligned}
2A'\phi' + A\phi'' &= 0, && (3.41)\\
\left(A^2\phi'\right)' &= 0. && (3.42)
\end{aligned}
$$

Eq. 3.41 is obviously and easy to solve, this imply $A^2\phi'$ must be a real constant

$$
\begin{aligned}
A^2\phi' &= C^2 && (3.43)\\
A &= \frac{C}{\sqrt{\phi'}}. && (3.44)
\end{aligned}
$$

However, Eq. 3.44 cannot be solved in general, hence we make a simple approximation. We assume that A varies slowly, so that the A'' term is negligible. More precisely, we should say that A''/A is much more smaller than both $\left(\phi'\right)^2$ and $p^2/\hbar^2$

$$\frac{A''}{A} = (\phi')^2 - \frac{p^2}{\hbar^2} \quad (3.45)$$

$$\frac{A''}{A} = 0, \quad (3.46)$$

and

$$(\phi')^2 = \frac{p^2}{\hbar^2} \quad (3.47)$$

$$\phi' = \pm\frac{p}{\hbar}. \quad (3.48)$$

Therefore, the function ϕ takes the form of

$$\phi = \pm\frac{1}{\hbar}\int p(x)\,dx. \quad (3.49)$$

Hence, the final form for the wavefunction is

$$\psi(x) = \frac{C}{\sqrt{\phi'}}e^{\pm\frac{i}{\hbar}\int p(x)dx}. \quad (3.50)$$

After obtaining the expression for the wavefunction, let us recall the expression as in Eq. 3.24 which is

$$\frac{\partial^2\Phi(p_x,p_y,z)}{\partial z^2} = \frac{2m}{\hbar^2}\left[E' + e\mathbf{F}\cdot\mathbf{z}\right]\Phi(p_x,p_y,z). \quad (3.51)$$

In order to solve the second order derivative, firstly we have to define the WKB wavefunction as the following expression

$$\Phi(p_x,p_y,z) = \frac{C}{\sqrt{p_z(z)}}\exp\left[\frac{i}{\hbar}S(p_x,p_y,z)\right], \quad (3.52)$$

with the momentum in z-direction

$$p_z(z) = \left|\frac{\partial S(p_x,p_y,z)}{\partial z}\right|. \quad (3.53)$$

The first task is to get the C constant, we take the initial value z_0

$$\Phi(p_x,p_y,z_0) = \frac{C}{\sqrt{p_z(z_0)}}\exp\left[\frac{i}{\hbar}S(p_x,p_y,z_0)\right], \quad (3.54)$$

and then the C constant becomes

$$C=\Phi\left(p_x,p_y,z_0\right)\sqrt{p_z\left(z_0\right)}\exp\left[-\frac{i}{\hbar}S\left(p_x,p_y,z_0\right)\right].$$

Next, we substitute into Eq. 3.52 and get

$$\Phi\left(p_x,p_y,z\right)=\Phi\left(p_x,p_y,z_0\right)\sqrt{\frac{p_z\left(z_0\right)}{p_z\left(z\right)}}\exp\left[\frac{i}{\hbar}S\left(p_x,p_y,z\right)-\frac{i}{\hbar}S\left(p_x,p_y,z_0\right)\right]. \tag{3.55}$$

Then, we perform the differentiation operation on the wavefunction $\Phi\left(p_x,p_y,z\right)$ and yield

$$\begin{aligned}\frac{\partial\Phi\left(p_x,p_y,z\right)}{\partial z} &= \frac{\partial}{\partial z}\left\{\Phi\left(p_x,p_y,z_0\right)\sqrt{p_z\left(z_0\right)}\exp\left[-\frac{i}{\hbar}S\left(p_x,p_y,z_0\right)\right]\frac{1}{\sqrt{p_z\left(z\right)}}\exp\left[\frac{i}{\hbar}S\left(p_x,p_y,z\right)\right]\right\}\\ &= \Phi\left(p_x,p_y,z_0\right)\sqrt{p_z\left(z_0\right)}\exp\left[-\frac{i}{\hbar}S\left(p_x,p_y,z_0\right)\right]\\ &\quad\times\frac{\partial}{\partial z}\left\{\frac{1}{\sqrt{p_z\left(z\right)}}\exp\left[\frac{i}{\hbar}S\left(p_x,p_y,z\right)\right]\right\}.\end{aligned} \tag{3.56}$$

After computing some complicated algebra, we obtain the solution for the second order derivative of $\Phi\left(p_x,p_y,z\right)$, which is shown as in Eq. 3.24

$$\frac{\partial^2\Phi\left(p_x,p_y,z\right)}{\partial z^2}=\frac{2m}{\hbar^2}\left[E'+e\mathbf{F}\cdot\mathbf{z}\right]\Phi\left(p_x,p_y,z\right), \tag{3.57}$$

with

$$E'=\frac{p_x^2}{2m}+\frac{p_y^2}{2m}+I_p. \tag{3.58}$$

Next, we perform some algebra manipulations as the following

$$\frac{1}{2}\left(\frac{\partial S\left(p_x,p_y,z\right)}{\partial z}\right)^2-Fz=E', \tag{3.59}$$

$$\left(\frac{\partial S\left(p_x,p_y,z\right)}{\partial z}\right)^2=2\left(E'+Fz\right), \tag{3.60}$$

$$p_z^2\left(z\right)=2\left(E'+Fz\right). \tag{3.61}$$

Eq. 3.59 is known as Hamilton-Jacobi equation(Salamin & Faisal, 1997) and the solution should be

$$S(p_x,p_y,z)-S(p_x,p_y,z_0)=\frac{1}{3F}\left(2E'+2Fz\right)^{\frac{3}{2}}-\frac{1}{3F}\left(2E'+2Fz_0\right)^{\frac{3}{2}}, \tag{3.62}$$

then,

$$\left[\frac{\partial S(p_x,p_y,z)}{\partial z}\right]^2 = 2m\left(E'+Fz\right), \tag{3.63}$$

$$\frac{\partial S(p_x,p_y,z)}{\partial z} = \sqrt{2m(E'+Fz)}. \tag{3.64}$$

By solving the integral of the action part,

$$\int \frac{\partial S(p_x,p_y,z)}{\partial z}dz=\int_{z_0}^{z}\left[2m\left(E'+Fz\right)\right]^{\frac{1}{2}}dz, \tag{3.65}$$

we get the final expression as

$$\begin{aligned} S(p_x,p_y,z)-S(p_x,p_y,z_0) &= \frac{1}{3mF}\left[2m\left(E'+Fz\right)\right]^{\frac{3}{2}}\Big|_{z_0}^{z} \qquad (3.66)\\ &= \frac{1}{3mF}\left(2mE'+2mFz\right)^{\frac{3}{2}}-\frac{1}{3mF}\left(2mE'+2mFz_0\right)^{\frac{3}{2}}. \end{aligned}$$

Next, we derive the action at exit point, where

$$\begin{aligned} z &= z_e \\ &= \frac{I_p}{F}, \end{aligned} \tag{3.67}$$

and then the action becomes

$$\begin{aligned} S(p_x,p_y,z_e)-S(p_x,p_y,z_0) &= \frac{1}{3mF}\left[\left(2m\left(\frac{p_x^2}{2m}+\frac{p_y^2}{2m}+I_p\right)+2mI_p\right)\right]^{\frac{3}{2}} \qquad (3.68)\\ &\quad -\frac{1}{3mF}\left(2mE'+2mFz_0\right)^{\frac{3}{2}} \\ &= \frac{1}{3mF}\left(p_x^2+p_y^2+4mI_p\right)^{\frac{3}{2}}-\frac{1}{3mF}\left(p_x^2+p_y^2+2mI_p+2mFz_0\right)^{\frac{3}{2}}. \end{aligned}$$

Let us set $p_\perp^2=p_x^2+p_y^2$, hence

$$
\begin{aligned}
S(p_x,p_y,z_e)-S(p_x,p_y,z_0) &= \frac{1}{3mF}\left(p_\perp^2+4mI_p\right)^{\frac{3}{2}}-\frac{1}{3mF}\left(p_\perp^2+2mI_p+2mFz_0\right)^{\frac{3}{2}} \\
&= \frac{1}{F}\left(I_p-Fz_0\right)\left(\sqrt{-2mI_p}\right)^3\left[1+\frac{3}{2}\left(p_\perp^2+6mI_p\right)\right] \\
&= \left(-i\kappa^3\right)\left(\frac{I_p}{F}-z_0\right)\left[1+\frac{3}{2}p_\perp^2+\frac{9}{2}\kappa\right], \qquad (3.69)
\end{aligned}
$$

and yields

$$
S(p_x,p_y,z)-S(p_x,p_y,z_0)=i\frac{\kappa^3}{3F}+i\frac{\kappa p_\perp^2}{2F}-i\kappa z_0. \qquad (3.70)
$$

As a result, we get

$$
\begin{aligned}
&\left[\frac{1}{3F}\left(\frac{p_x^2}{2m}+\frac{p_y^2}{2m}+I_p+Fz\right)\right]^{\frac{3}{2}}-\left[\frac{1}{3F}\left(\frac{p_x^2}{2m}+\frac{p_y^2}{2m}+I_p+Fz_0\right)\right]^{\frac{3}{2}} \qquad (3.71)\\
&= \frac{\sqrt{-2I_p}}{F}\left(\frac{2}{3}I_p+\frac{p_\perp^2}{2}-Fz_0\right).
\end{aligned}
$$

Next, we substitute Eq. 3.70 into Eq. 3.55 and yield

$$
\Phi(p_x,p_y,z)=\Phi(p_x,p_y,z_0)\sqrt{\frac{p_z(z_0)}{p_z(z)}}\exp\left[\frac{i}{\hbar}S(p_x,p_y,z)-\frac{i}{\hbar}S(p_x,p_y,z_0)\right], \qquad (3.72)
$$

imply that

$$
\begin{aligned}
\Phi(p_x,p_y,z) &= \Phi(p_x,p_y,z_0)\sqrt{\frac{p_z(z_0)}{p_z(z)}}\exp\left[\frac{i}{\hbar}\left(i\frac{\kappa^3}{3F}+i\frac{\kappa p_\perp^2}{2F}-i\kappa z_0\right)\right], \\
&= \Phi(p_x,p_y,z_0)\sqrt{\frac{p_z(z_0)}{p_z(z)}}\exp\left[\frac{1}{\hbar}\left(-\frac{\kappa^3}{3F}-\frac{\kappa p_\perp^2}{2F}+\kappa z_0\right)\right]. \qquad (3.73)
\end{aligned}
$$

Then, the tunnel ionization amplitude for a short range potential in a DC field is obtained

$$
\begin{aligned}
a_T(F,p_\perp) &= \sqrt{\frac{p_z(z_0)}{p_z(z)}}\exp\left[\frac{1}{\hbar}\left(-\frac{\kappa^3}{3F}-\frac{\kappa p_\perp^2}{2F}+\kappa z_0\right)\right] \qquad (3.74)\\
&= \sqrt{\frac{\kappa}{p_z(z)}}\exp\left[\frac{1}{\hbar}\left(-\frac{\kappa^3}{3F}-\frac{\kappa p_\perp^2}{2F}+\kappa z_0\right)\right].
\end{aligned}
$$

The momentum $p_z(z_0)$ is replaced with κ, since $z_0 \ll z_e$. Next, let us make a strong-field eikonal approximation and the resulting correction to the action is defined as below:

$$\Delta S = \int_{z_0}^{z} dz' \frac{V(z')}{p_z(z')}, \tag{3.75}$$

where the momentum is $p_z(z) = \sqrt{\kappa^2 - 2Fz}$.

When Coulomb potential is short range potential, $V(z) = \frac{Q}{z}$, then

$$\Delta S = \int_{z_0}^{z} dz \frac{Q}{z\sqrt{\kappa^2 - 2Fz}}. \tag{3.76}$$

Hence, the action takes the solution in the form of

$$\Delta S = \frac{Q}{\kappa} \ln\left(\frac{1+\sqrt{1-2Fz_0/\kappa^2}}{1-\sqrt{1-2Fz_0/\kappa^2}}\right). \tag{3.77}$$

Next, we set that the x takes the value of $2Fz_0/\kappa^2$, imply that

$$\begin{aligned} \Delta S &= \frac{Q}{\kappa} \ln\left(\frac{1+\sqrt{1-x}}{1-\sqrt{1-x}}\right) \\ &= \frac{Q}{\kappa} \ln\left(\frac{2\kappa^2}{Fz_0} - 2\right). \end{aligned} \tag{3.78}$$

In the work of (Murray, Liu, & Ivanov, 2010), they discuss that $\frac{F}{\kappa} \ll \kappa$ in the condition of $F \gg 1$, then the action is approximated as

$$\Delta S \approx \frac{Q}{\kappa} \ln\left(\frac{2\kappa^2}{Fz_0}\right). \tag{3.79}$$

Eqn 3.79 is known as the Coulomb correction as for the zero order contribution coresponding to the short-range potential. Hence, the Coulomb correction is included in a_T as in eqn 3.74 to get the final expression for the tunnel ionization amplitude of a hydrogen atom in a static field

$$a_T(F, p_\perp) = \sqrt{\frac{\kappa}{p_z(z)}} \left(\frac{2\kappa^2}{Fz_0}\right)^{Q/\kappa} \exp\left[\frac{1}{\hbar}\left(-\frac{\kappa^3}{3F} - \frac{\kappa p_\perp^2}{2F} + \kappa z_0\right)\right]. \tag{3.80}$$

This is the final result of the calculation of the tunneling amplitude. The only missing component is the bound wave function in the mixed coordinate-momentum representation.

3.2.1 Mixed representation for a bound wave function

1.) Mixed representation is obtained by simple application of the numerical Fourier transform with respect to the two dimensions orthogonal to the direction of tunneling.

2.) Analytical expression for the bound wave function in mixed representation (Sheehy et al., 1998).

The atomic wave function with quantum numbers l, m has a form of:

$$\Psi(x,y,z) = \frac{1}{\sqrt{2\pi}} e^{im\phi} \Psi_m(\rho,z), \tag{3.81}$$

where ϕ,ρ and z are the usual cylindrical coordinates.

Besides, eqn 3.81 does not include the polarization of the field-free bound state and also "unaware" of the modified potential barrier where the effect depend on z_0.

From eqn 3.14 and substitute the wavefunction inside:

$$\begin{aligned} \Phi(p_x,p_y,z_0) &= \frac{1}{2\pi}\int dx \int dy \Psi(x,y,z) \exp\left(\frac{-ixp_x - iyp_y}{\hbar}\right) \\ &= \frac{1}{2\pi}\int dx \int dy \frac{1}{\sqrt{2\pi}} e^{im\phi}\Psi_m(\rho,z) \exp\left(\frac{-ixp_x - iyp_y}{\hbar}\right) \\ &= \frac{1}{(2\pi)^{3/2}}\int dx \int dy \Psi_m(\rho,z) \exp\left(\frac{-ixp_x - iyp_y}{\hbar} + im\phi\right) \end{aligned} \tag{3.82}$$

Next, we transform equation 3.82 into polar coordinates and yield

$$\begin{aligned} \Phi(p_x,p_y,z_0) &= \frac{1}{(2\pi)^{3/2}}\int dx \int dy \Psi_m(\rho,z) \exp\left(\frac{-ixp_x - iyp_y}{\hbar}\right) e^{im\phi} \qquad (3.83) \\ &= \frac{e^{im\phi_0}}{(2\pi)^{3/2}} \int_0^\infty \rho d\rho \Psi_m(\rho,z_0) \int_0^{2\pi} d\phi \exp\left(\frac{-i\sqrt{p_x^2+p_y^2}\rho\cos\phi}{\hbar} + im\phi\right), \end{aligned}$$

following by

$$\Phi(p_x,p_y,z_0) = \frac{e^{im\phi_0}}{(2\pi)^{3/2}} \int_0^\infty \rho d\rho \Psi_m(\rho,z_0) \int_0^{2\pi} d\phi \exp\left(\frac{-ip_\perp\rho\cos\phi}{\hbar} + im\phi\right), \tag{3.84}$$

where $p_\perp$ is the perpendicular momentum $\sqrt{p_x^2 + p_y^2}$ and ϕ_0 is the angle of the perpendicular makes with the z axis.

Let us recall the identity of Bessel function which is defined as the following expression

$$J_m(x) = \frac{1}{2\pi}\int_0^{2\pi} \cos(m\phi - x\sin\phi)\, d\phi, \tag{3.85}$$

and another identity of Bessel function which is defined as

$$\exp\left[\left(\frac{x}{2}\right)\left(t - \frac{1}{t}\right)\right] = \sum_{n=-\infty}^{\infty} J_n(x)\, t^n. \tag{3.86}$$

Refer to our eqn, what we need is the representation of the form as the following expression

$$\int_0^{2\pi} \exp\left(\frac{-ip_\perp\rho\cos\phi}{\hbar} + im\phi\right) d\phi = \int_0^{2\pi} \exp\left(\frac{-ip_\perp\rho}{\hbar}\cos\phi\right) e^{im\phi} d\phi. \tag{3.87}$$

Hence, by making a substitution of

$$\exp\left[\left(\frac{x}{2}\right)\left(t - \frac{1}{t}\right)\right] = \exp\left(\frac{-ip_\perp\rho}{\hbar}\cos\phi\right), \tag{3.88}$$

then we can recover equation 3.84 into the Bessel function form

$$\begin{aligned}
\int_0^{2\pi} \exp\left(\frac{-ip_\perp\rho}{\hbar}\cos\phi\right) e^{im\phi} d\phi &= \int_0^{2\pi} \sum_{m=-\infty}^{\infty} J_m(x)\, t^m e^{im\phi} d\phi \\
&= \sum_{m=-\infty}^{\infty} J_m(x) \int_0^{2\pi} \left(-ie^{-i\phi}\right)^m e^{im\phi} d\phi \\
&= \sum_{m=-\infty}^{\infty} (-i)^m J_m(x) \int_0^{2\pi} d\phi \\
&= 2\pi(-i)^m J_m(x),
\end{aligned} \tag{3.89}$$

and subsequently we have

$$\Phi(p_x, p_y, z_0) = \frac{e^{im\phi_0}}{(2\pi)^{3/2}} \int_0^{\infty} \rho\, d\rho\, \Psi_m(\rho, z_0) \int_0^{2\pi} d\phi \exp\left(\frac{-ip_\perp\rho\cos\phi}{\hbar} + im\phi\right). \tag{3.90}$$

Hence, calculate the ϕ integral to obtain

$$\Phi\left(p_x, p_y, z_0\right) = \frac{e^{im\phi_0}}{(2\pi)^{1/2}} (-i)^m \int_0^\infty \rho d\rho \Psi_m\left(\rho, z_0\right) J_m\left(\frac{\rho p_\perp}{\hbar}\right). \tag{3.91}$$

When the value x is very small, the approximate formula for Bessel Function would be

$$J_n(x) = \frac{1}{\Gamma(n+1)}\left(\frac{x}{2}\right)^n + O\left(x^{n+2}\right), \tag{3.92}$$

imply that

$$\begin{aligned} J_m\left(\frac{\rho p_\perp}{\hbar}\right) &= \frac{1}{\Gamma(m+1)}\left(\frac{\rho p_\perp}{2\hbar}\right)^m + O\left(x^{m+2}\right) \\ &= \frac{1}{m!}\left(\frac{\rho p_\perp}{2\hbar}\right)^m. \end{aligned} \tag{3.93}$$

Hence, the exponential suppression of tunneling with nonzero $p_\perp$ (to replace the Bessel function with its limit for small arguments)

$$\Phi\left(p_x, p_y, z_0\right) = \frac{e^{im\phi_0}}{(2\pi)^{1/2}} \frac{(-i)^m}{m!} \int_0^\infty \rho d\rho \Psi_m\left(\rho, z_0\right)\left(\frac{\rho p_\perp}{2}\right)^m. \tag{3.94}$$

Let us define the asymtotic form of the hydrogen wave function

$$\Psi_{asymp}(x, y, z) = \frac{\kappa^{3/2} e^{im\phi}}{\sqrt{2\pi}} C_{\kappa l} N_{lm} (\kappa r)^{Q/\kappa - 1} e^{-\kappa r} P_l^m(\cos\theta), \tag{3.95}$$

where

$$C_{\kappa l} \frac{(-1)^{n-l-1} 2^n}{\sqrt{n(n+1)!(n-l-1)!}}, \tag{3.96}$$

$$N_{lm} = \sqrt{\frac{(2l+1)(l+m)!}{2(l-m)!}} \frac{1}{2^m m!}, \tag{3.97}$$

and

$$\kappa = \sqrt{2I_p}, \tag{3.98}$$

where I_p is the ionization potential.

In order to simplified Eq. 3.95, the asymtotic assumptions are

1. Legendre polynomial can be replaced with the limit for small angles,

$$P_l^m(\cos\theta) \propto \sin^m\theta \tag{3.99}$$

2. Asymtotic region where $\rho \ll z$ and $z > z_0$, imply that

$$\sin\theta \approx \frac{\rho}{z_0} \tag{3.100}$$

$$r \approx z_0 + \frac{\rho^2}{2z_0} \tag{3.101}$$

We insert Eq.3.95 into Eq.3.94 and yield,

$$\begin{aligned}\Phi(p_x, p_y, z_0) &= \frac{e^{im\phi_0}}{(2\pi)^{1/2}} \frac{(-i)^m}{m!} \int_0^\infty \rho d\rho \Psi_m(\rho, z_0) \left(\frac{\rho p_\perp}{2}\right)^m \qquad (3.102)\\ &= \frac{e^{im\phi_0}}{(2\pi)^{1/2}} \frac{(-i)^m}{m!} \int_0^\infty \rho d\rho \frac{\kappa^{3/2} e^{im\phi}}{\sqrt{2\pi}} C_{\kappa l} N_{lm} (\kappa r)^{Q/\kappa - 1} e^{-\kappa r} \left(\frac{\rho}{z_0}\right)^m \left(\frac{\rho p_\perp}{2}\right)^m\end{aligned}$$

following by

$$\begin{aligned}\Phi(p_x, p_y, z_0) &= \frac{(-i)^m e^{im\phi_0}}{(2\pi)^{1/2}} C_{\kappa l} N_{lm} \left(\frac{p_\perp}{2}\right)^m \frac{1}{m! z_0^m} \frac{\kappa^{3/2} e^{im\phi}}{\sqrt{2\pi}} (\kappa)^{Q/\kappa - 1} \qquad (3.103)\\ &\times \int_0^\infty d\rho\, (r)^{Q/\kappa - 1} e^{-\kappa r} \rho\, (\rho)^m (\rho)^m \\ &= \frac{(-i)^m e^{im\phi_0}}{(2\pi)^{1/2}} C_{\kappa l} N_{lm} \left(\frac{p_\perp}{2}\right)^m \frac{1}{m! z_0^m} \frac{\kappa^{3/2} e^{im\phi}}{\sqrt{2\pi}} (\kappa)^{Q/\kappa - 1} \qquad (3.104)\\ &\times \int_0^\infty d\rho \left(z_0 + \frac{\rho^2}{2z_0}\right)^{Q/\kappa - 1} e^{-\kappa\left(z_0 + \rho^2/2z_0\right)} \rho^{2m+1}\end{aligned}$$

Then, by applying a simple transformation on the integral and we get the following expression

$$\begin{aligned}\Phi(p_x, p_y, z_0) &= \frac{(-i)^m e^{im\phi_0}}{(2\pi)^{1/2}} C_{\kappa l} N_{lm} e^{-\kappa z_0} \left(\frac{p_\perp}{2}\right)^m \kappa^{Q/\kappa - 1/2} z_0^{Q/\kappa} \frac{1}{z_0} \frac{\kappa}{m! z_0^m} \frac{e^{im\phi}}{\sqrt{2\pi}} \int_0^\infty d\rho \rho^{2m+1} \\ &= \frac{(-i)^m e^{im\phi_0}}{(2\pi)^{1/2}} C_{\kappa l} N_{lm} e^{-\kappa z_0} \left(\frac{p_\perp}{2}\right)^m \kappa^{Q/\kappa - 1/2} z_0^{Q/\kappa} \qquad (3.105)\end{aligned}$$

Finally, we obtain the tunneling ionization amplitude which is

$$\Phi(p_x, p_y, z_0) = \frac{(-i)^m e^{im\phi_0}}{(2\pi)^{1/2}} C_{\kappa l} N_{lm} e^{-\kappa z_0} \left(\frac{p_\perp}{2}\right)^m \kappa^{Q/\kappa - 1/2} z_0^{Q/\kappa} \tag{3.106}$$

3.3 Multiphoton Ionization (MPI)

There exist an ionization process even though when the photons energy is less than the ionization potential. This phenomenon was first described by Maria Goeppert-Mayer about the idea of two-photon absoprtion "in one quantum act" effectively introduced the concept of a virtual absorption. Usually, for the absorption of a photon by an atom, one requires that the photon energy be equal to the difference in the energy levels between the ground state and a real excited state. If the photon energy does not match or "resonate" with a pair of stationary eigen-states of the atom, the absorption cannot occur.

Even if the real states do not exist for the resonance condition to be satisfied, the electron can for a very brief period of time (Kulander, 1987b) (as permitted by the uncertainty principle) absorb a photon temporarily and thus be excited to a "virtual state"- that is a state having the energy equal to the photon energy above the ground state of the atom.

If the field is strong enough to have a sufficient number of photons per unit volume, then the virtually excited electron may absorb another available photon even during the very short "lifetime" of the virtual state.

In principle, after one or more successive virtual absoprtions, the electron can always reach the continuous stationary (long-lived or real) eigen-states of the atom above the ionization threshold. Therefore, if the light field is strong enough, i.e. the number of density of photons is high enough, laser photons of any frequency and polarization can ionize the atom by successive intermediate virtual absorptions followed by the final transition to the real continuum states.

Some modifications from old Einsteinian photoelectric effect:

$$K.E. = n \times \text{ (photon energy)} - I_p, > 0, \text{ for any integer } n$$

first treated by Geoppert-Mayer using the second order perturbation theory, to the n-th order.

The lifetimes of the virtual states are typified by the difference-energy between a virtual state and the nearest stationary state.

When an atom is placed in an intense laser field (Krause, Schafer, & Kulander, 1992), there exists an interesting phenomenon where more than one photon is absorbed during the ionization process. It was first observed by ... For the N-photon case, the general expression of the transition rate for N number of photons from an initial state (i) to a final state (f) is

$$w_{i\to f}^{(N)} = \left| \sum_{ab\ldots yz} \frac{\mu_{ia}\mu_{ab}\ldots\mu_{yz}\mu_{zf}}{\hbar^N \left(\omega_{af}-(N-1)\,\omega\right)\left(\omega_{bf}-(N-2)\,\omega\right)\ldots\left(\omega_{yf}-2\omega\right)\left(\omega_{zf}-\omega\right)} E^N \right| \times 2\pi\rho_f\left(\omega_{if}-N\omega\right) \quad (3.107)$$

Let us consider n number of photons were absorbed during a nonresonant multiphoton ionization (Gribakin & Kuchiev, 1997). The celebrated ionization rate would be

$$\begin{aligned} w_n &\simeq (\sigma_1 I\tau)^{n-1}\sigma_1 I \quad (3.108) \\ &= \sigma_n I^n \end{aligned}$$

3.3.1 Resonance-Enhanced Multiphoton

For certain special case where the transition of the electron to a resonant state in the first hand then only ionized again from the particular excited state by absorbing a certain amount of photons again. For simplicity, let say the ground state electron absorbs k number of photons to be excited to a resonant electronic state, then by absorbing l amount of photons in order to be ionized from those resonant state. Hence, the total number of photons involve in this ionization process is $n = k + l$. Although the same number of photons are absorbed in the both multiphoton ionization (Kulander, 1987a), however the ionization rate for both processes are different.

Ordinary nonresonant MPI is limited by the lifetimes of the intermediate states (virtual states), if the system relaxes to its ground state before the absorption of the next photon, then the transition does not happen. Meanwhile, for the REMPI, the system is limited by the pulse duration of the laser, Δt_{pulse} and the ionization rate, w' for this process is given by

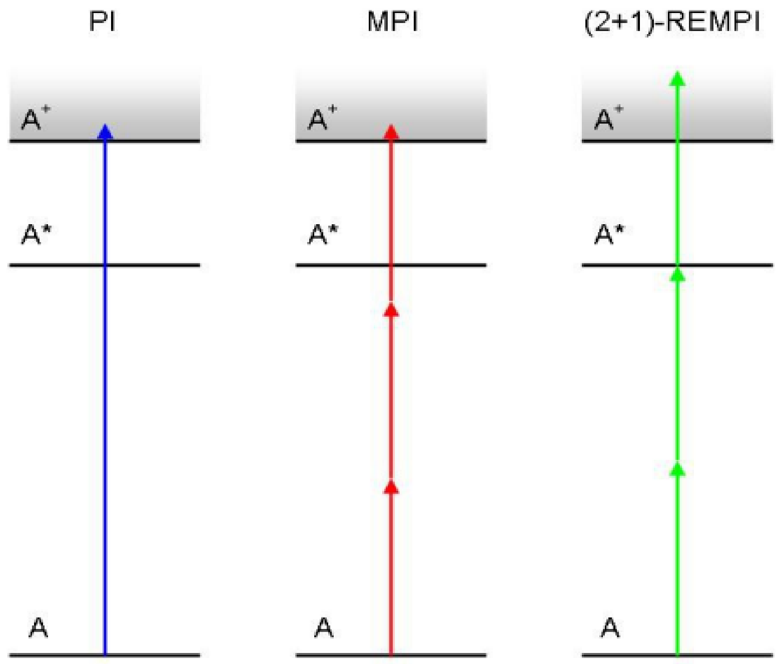


Figure 3.3: One-photon ionization (PI), non-resonant three-photon ionization (MPI), and resonance enhanced three-photon ionization ([2+1]-REMPI)

$$\begin{aligned} w'_n &\simeq \left\{(\sigma_1 I\tau)^{k-1}\sigma_1 I\right\}\Delta t_{pulse}\left\{(\sigma_1 I\tau)^{l-1}\sigma_1 I\right\} \\ &= w_n\frac{\Delta t_{pulse}}{\tau}. \end{aligned} \tag{3.109}$$

As what we can understand from the equation 3.109, the ionization rate for REMPI is greater than the ordinary MPI due to the factor of $\frac{\Delta t_{pulse}}{\tau}$. The main reason here is stationary state which is also known as eigenstate is having a greater lifetime than virtual states, as a result, the rate of REMPI is enhanced in comparison to the nonresonant MPI.

However, the enhancement is not applicable for all range of laser intensities. For a certain intensity which is high enough, the atom will reach the maximum ionization probability of 1 for both process. Let us define the MPI probability as

$$P = 1 - \exp\left(-\int_{-\infty}^{\infty}\sigma_N I^N dt\right), \tag{3.110}$$

where the I term is absorbing the spatial and temporal intensity distributions because same amount of photons are absorbed in both resonant and non-resonant cases. Therefore, we assume the probability only takes consideration of the single transition of the electron between the initial and final states, and thus generalize into two separate electronic transitions. Henceforth, the new probability in this REMPI would be

$$
\begin{aligned}
P &= P_{excitation} \times P_{ionization} && (3.111)\\
&= \left[1 - \exp\left(-\int_{-\infty}^{\infty} \sigma_N I^N dt\right)\right]\left[1 - \exp\left(-\int_{-\infty}^{\infty} \sigma_M I^M dt\right)\right] && (3.112)\\
&= \left(1 - e^{-\sigma_N I_0^N \tau}\right)\left(1 - e^{-\sigma_M I_0^M \tau}\right). && (3.113)
\end{aligned}
$$

3.4 Corkum's Model

In 1993, P. B. Corkum (Corkum, 1993) had introduced a model which is known as "The Simple Man Model" to explain the phenomenon where the electron is excited to the continuum state under the intense laser field, then recollide and recombine with the parent ion thus emit an amount of energy $I_p + 3.17U_p$ where U_p is the ponderomotive potential energy. These idea inspires the evolution of the high harmonic generation, which is a famous and pioneer research topic in the recent year.

In the dipole approximation, for a linearly polarized laser field, the interaction potential energy for an electron at position $\mathbf{r} = (x, y, z)$ is

$$V(\mathbf{r}, t) = V_{atom}(\mathbf{r}) + xeE_L(t), \quad (3.114)$$

where we assume that the laser electric field

$$E_L(t) = E_0 \cos(\omega_L t + \rho). \quad (3.115)$$

$E_L(t)$ is polarized along the x axis and positive when pointing in the direction of increasing x coordinate.The electron motion during the oscillation depends on the phase of the electric field $E_L(t)$ at which ionization has occured,

$$\phi_0 = \omega_L t_0 + \rho. \quad (3.116)$$

Consequently, the Coulomb interaction to the parent ion becomes a small perturbation compared to the laser field. The simplest approximation is to neglect the Coulomb field after ionization. Once, $V_{atom}(\mathbf{r})$ is dropped from equation 3.114, the center-of-mass motion of the quantum wave packet is described by

$$\begin{aligned} \ddot{x} &= -\frac{e}{m}E_L(t) && (3.117) \\ &= -\frac{e}{m}E_0\cos(\omega_L t+\rho). && (3.118) \end{aligned}$$

The vector potential $A_L(t)$ is defined by then following expression

$$E_L(t) = -\frac{dA_L}{dt}, \quad (3.119)$$

and hence

$$\begin{aligned} A_L(t) &= -\int E_L(t)\,dt && (3.120) \\ &= -\int E_0\cos(\omega_L t+\rho)\,dt \\ &= -\frac{eE_0}{m\omega_L}\sin(\omega_L t+\rho). && (3.121) \end{aligned}$$

This implies that the velocity of the electron:

$$\begin{aligned} v(t) &= A_L(t)-A_L(t_0) && (3.122) \\ &= -\frac{eE_0}{m\omega_L}\sin(\omega_L t+\rho)+\frac{eE_0}{m\omega_L}\sin(\omega_L t_0+\rho) && (3.123) \end{aligned}$$

and

$$\begin{aligned} v(t_f) &= v_0-\frac{eE_0}{m\omega_L}\sin(\omega_L t+\rho)+\frac{eE_0}{m\omega_L}\sin(\omega_L t_0+\rho) && (3.124) \\ &= v_0+\frac{eE_0}{m\omega_L}\left[\sin(\phi_0)-\sin(\phi_f)\right]. && (3.125) \end{aligned}$$

Since the velocity is the rate of changes in the propagation direction

$$\begin{aligned} \dot{x} &= -\frac{eE_0}{m\omega_L}\sin(\omega_L t+\rho)+\frac{eE_0}{m\omega_L}\sin(\omega_L t_0+\rho) && (3.126) \\ &= -\frac{eE_0}{m\omega_L}\sin\phi+\frac{eE_0}{m\omega_L}\sin\phi_0, && (3.127) \end{aligned}$$

then we can retrieve the position of the electron by

$$
\begin{aligned}
x &= \int_{t_0}^{t} v_0 - \frac{eE_0}{m\omega_L}\sin(\omega_L t+\rho) + \frac{eE_0}{m\omega_L}\sin(\omega_L t_0+\rho)\,dt' && (3.128)\\
&= v_0(t-t_0) + \left[\frac{eE_0}{m\omega_L^2}\cos(\omega_L t+\rho)\right]_{t_0}^{t} + \frac{eE_0}{m\omega_L}\sin(\omega_L t_0+\rho)(t-t_0) \\
&= \left[v_0 + \frac{eE_0}{m\omega_L}\sin(\omega_L t_0+\rho)\right](t-t_0) + \frac{eE_0}{m\omega_L^2}\left[\cos(\omega_L t+\rho) - \cos(\omega_L t_0+\rho)\right] \\
&= \left[v_0 + \frac{eE_0}{m\omega_L}\sin\phi_0\right]\frac{(\phi_f-\phi_0)}{\omega_L} + \frac{eE_0}{m\omega_L^2}\left[\cos\phi_f - \cos\phi_0\right], && (3.129)
\end{aligned}
$$

where ϕ's are the laser phases at $t=t_0$, $t=t_f$, $\phi_0=\omega_L t_0$ and $\phi_f=\omega t_f$.

From Eq. 3.124, one obtains a maximum final velocity

$$v_f = v_0 + 2\frac{eE_0}{m\omega_L}. \quad (3.130)$$

For the case where $\phi_0=\frac{\pi}{2}$ and $\phi_f=\frac{3\pi}{2}$, if $v_0=0$, the maximum energy E_f is

$$
\begin{aligned}
E_f &= \frac{mv_f^2}{2} && (3.131)\\
&= 2\frac{e^2E_0^2}{m\omega_L^2} && (3.132)\\
&= 8U_p && (3.133)
\end{aligned}
$$

where

$$
\begin{aligned}
U_p &= \frac{e^2E_0^2}{4m\omega_L^2} && (3.134)\\
&= \frac{I_o}{4\omega_L^2},
\end{aligned}
$$

U_p is called the ponderomotive energy. This maximum energy can only be reached at $x\neq 0$,

$$\left|x\left(t_f\right)\right| = \frac{E_0}{\omega^2}(2n-1)\pi, \quad n=1,2,... \quad (3.135)$$

If the electron initial velocity is $v_0=0$ at time t_0, then the maximum energy that the electron can acquire at its return to the parent ion, i.e. when $x\left(t_f\right)=0$ is $E_f=\frac{v_f^2}{2}=3.17U_p$.

3.5 Perelomov Popov Terent'ev (PPT) model

In 1966, Perelomov, Popov and Terent'ev (Perelomov, Popov, & Terent'ev, 1966) had formulated a theoretical model to explain the ionization rate of atom in laser field. This well known model is named after them which is known as the PPT model. To begin with this formalism, firstly the external electric field and subsequently the vector potential are given as the following expressions

$$\mathbf{E}(t) = \mathbf{E}\cos\omega t, \tag{3.136}$$

$$\mathbf{A}(t) = -\frac{\mathbf{E}}{\omega}\sin\omega t. \tag{3.137}$$

In the velocity gauge, $\mathbf{v} \to \mathbf{v}(t)$, we have

$$\begin{aligned} \mathbf{v}(t) &= \mathbf{p} + e\mathbf{A}(t) \\ &= \mathbf{p} - \frac{e\mathbf{E}}{\omega}\sin\omega t. \end{aligned} \tag{3.138}$$

We define the Volkov wavefunction, starting with time-dependent Schrodinger equation

$$i\hbar\frac{\partial}{\partial t}\Psi = \frac{\hbar^2}{2m}\left[\mathbf{p} + e\mathbf{A}(t)\right]^2\Psi. \tag{3.139}$$

$\mathbf{p}$ is canonical momentum, different from kinetic energy.

Let $\Psi = N\rho(t)\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)$

$$i\hbar\frac{\partial}{\partial t}\rho(t)\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) = \frac{\hbar^2}{2m}\left[\mathbf{p} + e\mathbf{A}(t)\right]^2\rho(t)\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) \tag{3.140}$$

$$\ln\rho(t) = \int_{t_0}^{t}\frac{\hbar^2}{2mi\hbar}\left[\mathbf{p} + e\mathbf{A}(t')\right]^2 dt'. \tag{3.141}$$

Then

$$\begin{aligned} \rho(t) &= \exp\left[\int_0^t -\frac{i\hbar}{2m}\left[\mathbf{p} + e\mathbf{A}(t')\right]^2 dt'\right] \\ &= \exp\left[\int_0^t -\frac{i\hbar}{2m}\left[p^2 t' + \frac{e^2\mathbf{E}^2}{2\omega^2}t + 2e\frac{\mathbf{p}\cdot\mathbf{E}}{\omega^2}\cos\omega t' - \frac{e^2\mathbf{E}^2}{8\omega^3}\sin 2\omega t'\right] dt'\right] \end{aligned} \tag{3.142}$$

In this case, we focus on the length gauge,

$$
\begin{aligned}
\Psi_{dE}(\mathbf{r},t) &= \exp\left(\frac{i}{\hbar}e\mathbf{A}(t)\cdot\mathbf{r}\right)N\rho(t)\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) \\
&= \frac{1}{\sqrt{(2\pi)^3}}\exp\left[\frac{i}{\hbar}(\mathbf{p}+e\mathbf{A}(t))\cdot\mathbf{r}\right]\exp\left[\int_0^t -\frac{i\hbar}{2m}\left[\mathbf{p}+e\mathbf{A}(t')\right]^2 dt'\right] \\
&= \frac{1}{\sqrt{(2\pi)^3}}\exp\left[\frac{i}{\hbar}\mathbf{v}(t)\cdot\mathbf{r}\right]\exp\left[\int_0^t -\frac{i\hbar}{2m}\mathbf{v}(t')^2 dt'\right]. \quad (3.143)
\end{aligned}
$$

The transition amplitude in time-dependent perturbation theory

$$
a_p(t) = -i\hbar\int_{t_i}^t dt'\left\langle\Psi_f^{(p)}\left|V_L(t')\right|\Psi_i\right\rangle\exp\left[\frac{i}{\hbar}\left(E_f-E_i\right)t\right], \quad (3.144)
$$

where $\Psi_f^{(p)}$ indicates the final state with momentum p at the detector and Ψ_i indicates the initial bound state.

However, in Keldysh case

$$
a_p(t) = -i\hbar\int_{t_i}^t dt'\left\langle\Psi_f^{(p)}\left|V_L(t')\right|\Psi_i\right\rangle\exp\left[\int_{t'}^t -\frac{i\hbar}{2m}\mathbf{v}(\tau)^2 d\tau\right]\exp\left[\frac{i}{\hbar}I_p t'\right]. \quad (3.145)
$$

This time dependent part represents the action

$$
\begin{aligned}
e^{iS(t)} &= \exp\left[\int_{t'}^t -\frac{i\hbar}{2m}\mathbf{v}(\tau)^2 d\tau\right]\exp\left[\frac{i}{\hbar}I_p t'\right], \quad (3.146) \\
S(t) &= \int_{t'}^t -\frac{i\hbar}{2m}\mathbf{v}(\tau)^2 d\tau + \frac{i}{\hbar}I_p t'. \quad (3.147)
\end{aligned}
$$

Let us define

$$
\omega t_0'' = \sinh^{-1}\gamma, \quad (3.148)
$$

then the action becomes

$$
S(t,t') = -\int_{t'}^t \frac{i\hbar}{2m}\mathbf{v}(\tau)^2 d\tau + \frac{i}{\hbar}I_p t'. \quad (3.149)
$$

We can see that only the imaginary part of action contributes to the decay rate which is shown by

$$
ImS(0,it_0'') = \int_{t'}^t \frac{\hbar}{2m}\mathbf{v}(i\tau)^2 d\tau - \frac{1}{\hbar}I_p t_0''. \quad (3.150)
$$

Since the momentum

$$\begin{aligned} m\mathbf{v}(t) &= \mathbf{p}+e\mathbf{A}(t) \\ &= \mathbf{p}-\frac{e\mathbf{E}}{\omega}\sin\omega t, \end{aligned} \tag{3.151}$$

then we have after defining an imaginary time $t \to it$

$$\begin{aligned} m\mathbf{v}_1(it) &= \mathbf{p}+e\mathbf{A}(it) \\ &= \mathbf{p}-i\frac{e\mathbf{E}}{\omega}\sinh\omega t. \end{aligned} \tag{3.152}$$

The ponderomotive potential part of the solution

$$\frac{\hbar}{2m}\int_{t_0''}^{t}\mathbf{v}(i\tau)^2\,d\tau = \frac{\hbar}{2m}\int_{t_0''}^{t}\mathbf{v}(i\tau)^2\,d\tau \tag{3.153}$$

$$= \frac{\hbar}{2m}\left[\frac{e^2E^2}{2\omega^2}\frac{\sinh\left(2\omega t_0''\right)}{2\omega}-\frac{e^2E^2}{2\omega^2}t_0''\right]$$

$$= \frac{\hbar}{m}\left[\frac{e^2E^2}{4\omega^2}\frac{2\gamma\sqrt{1+\gamma^2}}{2\omega}-\frac{e^2E^2}{4\omega^2}t_0''\right], \tag{3.154}$$

and

$$\begin{aligned} \sinh\left(2\omega t_0''\right) &= 2\sinh\left(\omega t_0''\right)\cosh\left(\omega t_0''\right) \\ &= 2\gamma\sqrt{1+\gamma^2}. \end{aligned} \tag{3.155}$$

Since we have the relation between Keldysh parameter γ and $\omega t_0''$

$$\omega t_0'' = \sinh^{-1}\gamma, \tag{3.156}$$

this implies that

$$\begin{aligned} ImS\left(0,it_0''\right) &= \int_{t'}^{t}\frac{\hbar}{2m}\mathbf{v}(i\tau)^2\,d\tau-\frac{1}{\hbar}I_pt_0'' \\ &= \frac{\hbar}{2m}\left[\frac{e^2E^2}{4\omega^2}\frac{2\gamma\sqrt{1+\gamma^2}}{2\omega}-\frac{e^2E^2}{4\omega^2}t_0''\right]-\frac{1}{\hbar}I_pt_0'' \\ &= -\frac{I_p}{\hbar\omega}\left[\left(1+\frac{\hbar^2}{2\gamma^2}\right)\sinh^{-1}\gamma-\frac{\hbar^2\sqrt{1+\gamma^2}}{2\gamma}\right] \end{aligned} \tag{3.157}$$

where the Keldysh parameter, γ takes the form of

$$\gamma = \frac{\sqrt{2mI_p}\omega}{eE} \tag{3.158}$$

Hence,

$$ImS\left(0, it_0''\right) = -\frac{I_p}{\hbar\omega}\left[\left(1+\frac{\hbar^2}{2\gamma^2}\right)\sinh^{-1}\gamma - \frac{\hbar^2\sqrt{1+\gamma^2}}{2\gamma}\right] \tag{3.159}$$

For the first case where $\gamma \ll 1$, then we have $\sinh^{-1}\gamma \approx \gamma$ meanwhile for the case $\gamma \gg 1$, we have $\sinh^{-1}\gamma \approx \ln 2\gamma$. Henceforth, when the Keldysh parameter $\gamma \ll 1$, the exponential function of Eq. 3.157 would be

$$\begin{aligned}
\exp\left[ImS\left(0, it_0''\right)\right] &= \exp\left\{-\frac{I_p}{\hbar\omega}\left[\left(1+\frac{\hbar^2}{2\gamma^2}\right)\sinh^{-1}\gamma - \frac{\hbar^2\sqrt{1+\gamma^2}}{2\gamma}\right]\right\} \\
&= \exp\left\{-\frac{I_p}{\hbar\omega}\left[\left(\gamma+\frac{\hbar^2}{2\gamma^2}\right) - \frac{\hbar^2\sqrt{1+\gamma^2}}{2\gamma}\right]\right\} \\
&= \exp\left[-\frac{\left(2I_p\right)^{3/2}}{3F}\right],
\end{aligned} \tag{3.160}$$

Then, when $\gamma \gg 1$, we have

$$\begin{aligned}
\exp\left[ImS\left(0, it_0''\right)\right] &= \exp\left\{-\frac{I_p}{\hbar\omega}\left[\left(1+\frac{\hbar^2}{2\gamma^2}\right)\sinh^{-1}\gamma - \frac{\hbar^2\sqrt{1+\gamma^2}}{2\gamma}\right]\right\} \\
&= \left(\frac{1}{2\gamma}\right)^{I_p/\hbar\omega} \\
&\approx E^{I_p/\hbar\omega}.
\end{aligned} \tag{3.161}$$

Note for the propagation operator : Evolution operator propagates vector from t_0 to t and it can be described by

$$|k,t\rangle = U\left(t, t_0\right)|k,t\rangle, \tag{3.162}$$

where the propagation operator is defined by

$$\begin{aligned}
U_0\left(t, t_0\right) &= e^{-i\mathbf{H}_0\left(t-t_0\right)} \\
&= \sum_n |n\rangle e^{-iE_n\left(t-t_0\right)}\langle n|.
\end{aligned} \tag{3.163}$$

As a result, this imply that

$$|k,t\rangle = \sum_n |n\rangle e^{-iE_n(t-t_0)} \langle n|k,t_0\rangle. \tag{3.164}$$

Time-evolution problem is solved once initial vector is expanded in terms of eigenvectors of stationary problem

$$i\hbar\frac{\partial}{\partial t}\Psi = H_0\Psi \tag{3.165}$$

with

$$\Psi(t_0) = \Psi_0 \tag{3.166}$$

$$H_0\rho_n = E_n\rho_n \tag{3.167}$$

Hence, we can rewrite the wavefunction as the summation of all states

$$\Psi(x,t) = \sum_n \rho_n(x) e^{-iE_n(t-t_0)} \langle \rho_n|\Psi_0\rangle, \tag{3.168}$$

and yield

$$\langle x|k,t\rangle = \sum_n \int dx' \langle x|n\rangle e^{-iE_n(t-t_0)} \langle n|x'\rangle \langle x'|k,t_0\rangle. \tag{3.169}$$

Next, let us rewrite again for the expression of the wavefunction in term of Green function in one dimension, along x

$$\Psi\left(x'',t\right) = \int dx' G\left(x'',t;x',t_0\right) \Psi_0\left(x',t_0\right) \tag{3.170}$$

where

$$G\left(x'',t;x',t_0\right) = \sum_n \langle x''|n\rangle \langle n|x'\rangle e^{-iE_n(t-t_0)} \tag{3.171}$$

Hence, for the propagation of an electron in the laser field, let us reconstruct the Volkov wavefunction as a basis

$$\begin{aligned}\Psi_{dE}(\mathbf{r},t) &= \frac{1}{\sqrt{(2\pi)^3}}\exp\left[\frac{i}{\hbar}(\mathbf{p}+e\mathbf{A}(t))\cdot\mathbf{r}\right]\exp\left[\int_0^t -\frac{i\hbar}{2m}\left[\mathbf{p}+e\mathbf{A}(t')\right]^2 dt'\right] \\ &= \frac{1}{\sqrt{(2\pi)^3}}\exp\left[\frac{i}{\hbar}\mathbf{v}(t)\cdot\mathbf{r}\right]\exp\left[\int_0^t -\frac{i\hbar}{2m}\mathbf{v}(t')^2 dt'\right] \qquad (3.172)\end{aligned}$$

In three dimensional space, the Green function propagator would be

$$G(\mathbf{r}'',t;\mathbf{r}',t') = \frac{\theta(t-t')}{(2\pi)^3}\int d^3\mathbf{p}\exp\left[\frac{i}{\hbar}\left(\mathbf{v}(t)\cdot\mathbf{r}-\mathbf{v}(t')\cdot\mathbf{r}'\right)\right]\exp\left[-\frac{i\hbar}{2m}\int_{t'}^t \mathbf{v}(\tau)^2 d\tau\right] \tag{3.173}$$

and we obtain the final expression for ionization rate

$$w = C_{n^*,l^*}^2\left(\frac{3\xi}{\pi\xi_0}\right)^{\frac{1}{2}} E\frac{(2l+1)(l+|m|)!}{2^{|m|}(|m|)!(l-|m|)!}\left(\frac{2\xi_0}{\xi}\right)^{2n-|m|-1}\exp\left(-\frac{2\xi_0}{3\xi}\right) \tag{3.174}$$

where

$$n^* = Z(2E)^{-1/2} \tag{3.175}$$

$$\xi_0 = (2E)^{1/2} \tag{3.176}$$

CHAPTER 4

PERTURBATIVE SEMI-ANALYTICAL (KELDYSH TYPE) THEORY

4.1 Introduction

Over the past century, there exist several models to describe the photoionization rate of an atom in an intense laser field, such as Smirnov and Chibisov's model, ADK (Ammosov, Delone and Krainov) theory, PPT (Perelomov, Popov and Terent'ev) and Keldysh's theory. In this chapter, the Keldysh's formalism is introduced where it describes the transition rate of an electron from ground state to Volkov state when an atom is placed in an intense laser field. The pioneering work of Leonid Keldysh was first introduced in 1965 which provides a complete theoretical description of the tunnel ionization by intense linearly polarized light where the photon energy, $\hbar\omega$ is lower than the ionization potential, I_p. In this chapter, detail derivation on the Keldysh's formalism and the extension of the model is outlined. We will discuss the output and the comparison of both model.

4.2 Keldysh's Ionization Rate in Linear Field

In variable electric field which is defined by the equation below,

$$E(t) = E\cos\omega t, \tag{4.1}$$

and the adiabatic parameter, γ

$$\gamma = \frac{\omega}{\omega_t} \tag{4.2}$$

$$= \frac{\omega\sqrt{2mI}}{eE} \tag{4.3}$$

$$= \frac{1}{2K_0F}. \tag{4.4}$$

On the other hand, γ is also known as the ratio between frequency of laser light, ω and frequency of electron tunneling, ω_t through a potential barrier where I is the ionization potential of atomic level.

$$I = \frac{\kappa^2 me^4}{2\hbar^2}, \tag{4.5}$$

while E is the amplitude of electric wave field and F is the reduced field

$$F = \frac{E}{\kappa^3 \varepsilon_a} \tag{4.6}$$

The multiquantumness parameter of the process is given by K_0 i.e. the minimal number of photons required for ionization,

$$K_0 = \frac{I}{\hbar\omega}. \tag{4.7}$$

From here, we note that

$$\kappa = \sqrt{\frac{I}{I_H}}. \tag{4.8}$$

There are some important points to take note, which is F, K_0 and γ are dimensionless quantities. Here, I_H is the ionization potential of the hydrogen atom,

$$I_H = \frac{me^4}{2\hbar^2} = 13.6eV, \tag{4.9}$$

and ε_a is the atomic unit of electric field intensity

$$\varepsilon_a = \frac{m^3 e^5}{\hbar^4} = 5.14 \times 10^9 \text{ Vcm}^{-1} \tag{4.10}$$

and the ionization rate w of a level is measured in the unit

$$w = \frac{me^4}{\hbar^3} = 4.13 \times 10^{16} \text{ s}^{-1} \tag{4.11}$$

Tunnel ionization take place when $\gamma \ll 1$, while for $\gamma \gg 1$ the ionization is a multiphoton process. We will discuss these two important processes in the following section where the Keldysh's formalism will be presented in details.

For a linearly polarized monochromatic electromagnetic wave, the differential ionization probability , i.e the momentum photoelectron spectrum , is of the form,

$$dw(\mathbf{p}) = P\exp\left\{-2K_0\left[f(\gamma) + c_1(\gamma)q_\parallel^2 + c_2(\gamma)q_\perp^2\right]\right\}\frac{d^3p}{(2\pi)^3}, \tag{4.12}$$

where $q{=}p/\kappa$ and $f(\gamma)$ is the keldysh function

$$f(\gamma) = \left(1+\frac{1}{2\gamma^2}\right)\operatorname{arcsin}\gamma - \frac{\sqrt{1+\gamma^2}}{2\gamma} \tag{4.13}$$

$$= \left\{ \begin{array}{l} \frac{2}{3}\gamma - \frac{1}{15}\gamma^3 \ , \ \gamma \ll 1 \\ \ln 2\gamma - \frac{1}{2} \quad , \ \gamma \gg 1 \end{array} \right\}.$$

The coefficients of the photoelectron momentum distribution are

$$c_1(\gamma) = \operatorname{arcsin}\gamma - \gamma(1+\gamma^2)^{-1/2} \tag{4.14}$$

$$c_2(\gamma) = \operatorname{arcsin}\gamma, \tag{4.15}$$

and $P(\gamma)$ is the pre-exponential factor while the definition for $\operatorname{arcsin}\gamma$ is

$$\operatorname{arcsin}\gamma \equiv \ln(\gamma + \sqrt{1+\gamma^2}. \tag{4.16}$$

Note that $\mathbf{p} = (p_{\|}, p_{\perp})$ is the photoelectron momentum, with $p_{\|}$ being the momentum component along the direction of the electric field E , $p_{\perp}$ being perpendicular to it and $\kappa = \sqrt{2I}$ being the characteristic momentum of the bound state.

For the ionization rate of a level (i.e , the probability of ionization per unit time) we have with an exponential accuracy

$$w(F,\omega) \left\{ \begin{array}{lr} \exp\left\{-\frac{2}{3F}\left[1-\frac{1}{10}\left(1-\frac{1}{3}\xi^2\right)\gamma^2\right]\right\} \ , & \gamma \ll 1 \\ (K_0F)^{2K_0} \sim J^{K_0} \quad , & \gamma \gg 1 \end{array} \right\}, \tag{4.17}$$

where J is the intensity of laser radiation and ξ is the elilipticity $[\xi^2 1]$ expressed by the following expression

$$J = \left(\frac{c}{8\pi}\right)\left(1+\xi^2\right)E^2 \tag{4.18}$$

For $\gamma \ll 1$ the ionization rate of a state $|lm\rangle$ with the orbital abgular momentum l by linearly polarized light ($\xi = 0$) is

$$w_{lm} = \kappa^2 \sqrt{\frac{3}{\pi}}(2l+1)\frac{(l+m)!}{2^m m!(l-m)!}C_{\kappa l}^2 2^{2n^*-m} \times F^{m+1.5-2n^*} \exp\left[-\frac{2}{3F}\left(1-\frac{1}{10}\gamma^2\right)\right] \tag{4.19}$$

with $w_{l,-m} = w_{lm}$ and $m0$.

From here, we can understand that $m = 0, \pm 1, ...$is the projection of the angular momentum l on the electric field and n^* is the effective principal quantum number of the level which is calculated from the experimentally measured energy $E_0 = -I$ of the atomic state:

$$n^* = \frac{Z}{\kappa} = \frac{Z}{\sqrt{2I}}, \tag{4.20}$$

where Z is the atomic or ion core charge, and $C_{\kappa l}$ is the dimensionless asymptotic coefficient of the atom wave function away ($\kappa r \gg 1$) from the nucleus.

The sufficiently precise expression for this coefficient is

$$C_{\kappa l}^2 = \frac{2^{2n^*-2}}{n^*(n^*+l)!(n^*-l-1)} \quad , \quad x! \equiv \Gamma(x+1) \tag{4.21}$$

The equation of ionazation rate $w_{lm}$is valid for low -frequency laser radiation, i.e. for $\omega \ll \omega_t$.

For an arbitrary γ , we obtain the final expression of the rate of ionization for the s level bound by a short-range ($Z = 0$) potential is represented in the form of the sum of n-photon process probabilities:

$$w(\varepsilon, \omega) = \sum_{n > n_{th}} w_n \quad , \quad n_{th} = K_0 (1 + \frac{1}{2\gamma^2}), \tag{4.22}$$

where $l = 0$, w_n is the partial probability of n-photon ionization:

$$w_n = \frac{\kappa^2}{\pi} |C_\kappa|^2 K_0^{-3/2} \beta^{1/2} F(\sqrt{\beta(n - n_{th})}) \times \exp\left\{ -\left[\frac{2}{3F} g(\gamma) + 2c_1(n - n_{th}) \right] \right\} \tag{4.23}$$

with

$$g(\gamma) = \frac{3f(\gamma)}{2\gamma}, \tag{4.24}$$

$$\begin{aligned} f(\gamma) &= \left(1 + \frac{1}{2\gamma^2}\right) \arcsin\gamma - \frac{\sqrt{1+\gamma^2}}{2\gamma} \\ &= \left\{ \begin{array}{l} \frac{2}{3}\gamma - \frac{1}{15}\gamma^3 \ , \ \gamma \ll 1 \\ \ln 2\gamma - \frac{1}{2} \quad , \ \gamma \gg 1 \end{array} \right\}, \end{aligned} \tag{4.25}$$

$$c_1(\gamma) = \arcsin\gamma - \gamma(1+\gamma^2)^{-1/2}. \tag{4.26}$$

The value n_{th} is the photoionization threshold for linearly polarized radiation, and the β value is

$$\begin{aligned} \beta &= 2(c_2 - c_1) \\ &= \frac{2\gamma}{\sqrt{1+\gamma^2}}, \end{aligned} \tag{4.27}$$

and the function F is defined as

$$\begin{aligned} F(x) &= \int_0^x \exp\left[-(x^2 - y^2)\right] dy \\ &= \left\{ \begin{array}{ccc} x - \frac{2}{3}x^2 + ... & , & x \longrightarrow 0 \\ \frac{1}{2x} + \frac{1}{4x^3+} + ... & , & x \longrightarrow \infty \end{array} \right\}. \end{aligned} \tag{4.28}$$

4.2.1 The Keldysh Function

The frequency dependence of the ionization rate of an atom is determined primarily by the function $f(\gamma, \xi)$. This function was calculated by L.V Keldysh (1964) for $\xi = 0$ which referred to as the Keldysh function.

For $\xi = 0$ we have

$$f(\gamma) = \sum_{n=0}^{\infty} (-1)^n f_n \gamma^{2n+1}, \tag{4.29}$$

and

$$f_n = \frac{2}{3} g_n = \frac{(2n-1)!!}{m! 2^{n-1} (2n+1)(2n+3)}, \tag{4.30}$$

which is a similar series for coefficients of the momentum spectrum $c_{1,2}(\gamma)$.

Since $f_n \propto n^{-5/2}$ for $n \longrightarrow \infty$, the series converge for $|\gamma| 1$. In the antiadiabatic domain, Eq. 4.29 becomes

$$f(\gamma) = \left(1 + \frac{1}{2\gamma^2}\right) \ln\gamma + \sum_{n=0}^{\infty} a_n \gamma^{-2n} \quad \text{as} \quad \gamma \longrightarrow \infty \tag{4.31}$$

where

$$
\begin{aligned}
a_0 &= \ln 2 - 1/2, & (4.32)\\
a_1 &= \ln 2/2, & (4.33)\\
a_2 &= 3/32, & (4.34)\\
a_3 &= -5/192, & (4.35)
\end{aligned}
$$

and so on.

Then, Eq. 4.12 becomes

$$dw(\mathbf{p}) = P\exp\left\{-2K_0\left[f(\gamma)+c_1(\gamma)q_{\parallel}^2+c_2(\gamma)q_{\perp}^2\right]\right\}\frac{d^3p}{(2\pi)^3}, \qquad (4.36)$$

which can be shown to remain valid in the case of linear polarization, with

$$f(\gamma) = \int_0^{\gamma}\chi(u)\left(1-\frac{u^2}{\gamma^2}\right)du, \qquad (4.37)$$

and

$$
\begin{aligned}
c_1(\gamma) &= c_2 - \gamma c_2' = \int_0^{\gamma}\left[\chi(u)-\chi(\gamma)\right]du, & (4.38)\\
c_2(\gamma) &= \int_0^{\gamma}\chi(u)du, & (4.39)
\end{aligned}
$$

where the function $\chi(u)$ is being completely defined by the shape of the laser pulse.

In the case where the external field is spatially uniform and is linearly polarized,

$$E(t) = E\varphi(\omega t)\ , \quad -\infty t\infty \quad , \quad \varphi(\pm\infty)\longrightarrow 0 \qquad (4.40)$$

It is possible to suggest a simple analytical procedure for determining $\chi(u)$ from the pulse shape. For instance, $\chi(u) = (1+u^2)^{-1/2}$ correspond to the monochromatic laser light with $\varphi(t) = \cos t$, $\chi(u) = 1/(1+u^2)$ to a soliton-like pulse with $\varphi(t) = 1/\cosh^2 t$.,etc.

When $\chi(u)$ is known in the analytical form, from expression

$$f(\gamma) = \int_0^{\gamma}\chi(u)\left(1-\frac{u^2}{\gamma^2}\right)du. \qquad (4.41)$$

It is easy to obtain adiabatic expansion. In particular, by setting

$$\chi(u) = (1+u^2)^{-\rho}, \qquad (4.42)$$

and hence, we obtain

$$f(\gamma)=\frac{2}{3}\gamma_2F_1\left(\frac{1}{2},\rho;\frac{5}{2},-\gamma^2\right), \tag{4.43}$$

$$f_n=\frac{2\Gamma(n+\rho)}{n!(2n+1)(2n+3)\Gamma(\rho)}\propto n^{\rho-3}, \tag{4.44}$$

where ρ is the ellipticity of light andfor $\gamma\longrightarrow\infty$

$$f(\gamma)\longrightarrow\frac{\sqrt{\pi}\Gamma(\rho-1/2)}{2\Gamma(\rho)},\qquad \rho>\frac{1}{2} \tag{4.45}$$

Meanwhile for $\rho=1/2$ the function $f(\gamma)$ grows as $\ln\gamma$. Then the shape of the field pulse corresponding to formula

$$\chi(u)=(1+u^2)^{-\rho}, \tag{4.46}$$

is characterized by the asymptotics

$$\varphi(t)=1-\rho t^2+\frac{1}{6}(7\rho^2-3\rho)t^4+......,\qquad t\longrightarrow 0, \tag{4.47}$$

$$\varphi(t)\approx\left\{\begin{array}{ll}[2(\rho-1)t]^{-\rho/(\rho-1)} & ,\ \rho>1,\\ 4\exp(-2t) & ,\ \rho=1\end{array}\right\}, \tag{4.48}$$

with $\varphi(t)=\cos t$, $1/\cosh^2 t$, and $(1+t^2)^{-3/2}$ correspondin to the values of $\rho=1/2,1,$and $3/2$, respectively.

For an arbitrary $\varphi(t)$ we have the expension

$$\chi(u)=1-\frac{1}{2}a_2u^2+\frac{5}{12}(a_2^2-0.1a_4)u^4-\frac{7}{18}\left(a_2^3-\frac{1}{5}a_2a_4+\frac{1}{280}a_6\right)u^6+... \tag{4.49}$$

which give, upon substitution into expression

$$f(\gamma)=\int_0^{\gamma}\chi(u)\left(1-\frac{u^2}{\gamma^2}\right)du. \tag{4.50}$$

The expansion of $g(\gamma)$ and the coefficients of the momentum spectrum $b_{1,2}(\gamma)$ in the adiabatic domain. From here, a_n are the coefficients of the series

$$\varphi(t)=1-\frac{a_2}{2!}t^2+\frac{a_4}{4!}t^4-......,\qquad a_2>0 \tag{4.51}$$

The Keldysh function for the case of linear polarization can be written in the form

$$f(\gamma,0)=\tau_0-\frac{1}{4\gamma^2}(\sinh 2\tau_0-2\tau_0) \tag{4.52}$$

where $\tau_0=arcsinh\ \gamma$.

4.3 Atom in linear polarized intense laser field

Attempting to calculate the photoionization rate is prohibitively complicated if done in a rigidly formal sense. In this section, we will study the interaction of a hydrogen atom with the intense linear polarized laser field in z-direction, which is described by the following equation

$$\mathbf{E} = E\cos\omega t\widehat{\mathbf{z}} \tag{4.53}$$

where E defines the electric field strength, ω is the frequency of the intense laser and $\widehat{\mathbf{z}}$ represents the unit vector in z-direction. The derivation is consisted of two main parts which is:

1.)Matrix element prefactor : An integral that represents the transition of an electron from hydrogen ground state to continuum state.

2.)Action part : The integral over time that show the action process for the transition of electron from hydrogen ground state to continuum state.

Next, we will introduce the Volkov wavefunction in details where it plays an important role in the Keldysh's formalism.

4.3.1 Volkov wavefunction

The Volkov wavefunction is defined as the following expression

$$\psi_p(\mathbf{r},t) = \exp\left\{\frac{i}{\hbar}\left[\Pi(t)\cdot\mathbf{r} - \frac{1}{2m}\int_0^t\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}\sin\omega\tau\right)^2 d\tau\right]\right\} \tag{4.54}$$

where it describe the free electron in the continuum state after the interaction with an intense laser field.

From here, we will show how Eq. 4.54 is derived. In an external electric field

$$\mathbf{E} = E\cos\omega t\widehat{\mathbf{z}}. \tag{4.55}$$

We recall the Maxwell equations as define in the following expressions,

$$\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t} - \nabla\phi, \tag{4.56}$$

$$\mathbf{B} = \nabla\times\mathbf{A}. \tag{4.57}$$

Note that if $\mathbf{A}$ is tranverse vector potential, implies that coulomb gauge $\nabla \cdot \mathbf{A} = 0$. Hence,

$$\nabla \cdot \mathbf{E} = \frac{\partial}{\partial t} \nabla \cdot \mathbf{A} - \nabla^2 \phi \tag{4.58}$$

then,

$$\mathbf{E}_T = -\frac{\partial \mathbf{A}}{\partial t} \tag{4.59}$$

$\mathbf{E}$ is the external electric field which is depending on the travelling time t at fix direction $\widehat{\mathbf{z}}$, hence,

$$\begin{aligned} \mathbf{A} &= -\int \mathbf{E}_T \, dt && (4.60) \\ &= -\int \mathbf{E} \cos \omega t \, dt \\ &= -\frac{\mathbf{E}}{\omega} \sin \omega t && (4.61) \end{aligned}$$

Next, we substitute the vector potential $\mathbf{A}$ into the Schrödinger's equation and yield,

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \Psi(\mathbf{r}, t) &= \frac{1}{2m} \left(\frac{\hbar}{i} \nabla - \frac{e}{c} \mathbf{A} \right)^2 \Psi(\mathbf{r}, t) \\ &= \frac{1}{2m} \left(\frac{\hbar}{i} \nabla + \frac{e\mathbf{E}}{\omega} \sin \omega t \right)^2 \Psi(\mathbf{r}, t). && (4.62) \end{aligned}$$

Then, by using separation of variables, let us define the wavefunction as

$$\Psi(\mathbf{r}, t) = \exp\left(\frac{i\mathbf{p} \cdot \mathbf{r}}{\hbar} \right) f(t), \tag{4.63}$$

and the Schrödinger's equation is expanded

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \exp\left(\frac{i\mathbf{p} \cdot \mathbf{r}}{\hbar} \right) f(t) &= \frac{1}{2m} \left(\frac{\hbar}{i} \nabla + \frac{e\mathbf{E}}{\omega} \sin \omega t \right)^2 \exp\left(\frac{i\mathbf{p} \cdot \mathbf{r}}{\hbar} \right) f(t) && (4.64) \\ &= \frac{1}{2m} \left\{ -\hbar^2 \nabla^2 + \left[\frac{e\mathbf{E}}{\omega} \right]^2 \sin^2 \omega t + \frac{\hbar}{i} \nabla \cdot \frac{e\mathbf{E}}{\omega} \sin \omega t + \frac{\hbar}{i} \frac{e\mathbf{E}}{\omega} \sin \omega t \cdot \nabla \right\} \\ &\quad \times \exp\left(\frac{i\mathbf{p} \cdot \mathbf{r}}{\hbar} \right) f(t). && (4.65) \end{aligned}$$

In the coulomb gauge, we know that the dot product of the vector potential is

$$\nabla \cdot \mathbf{A} = 0 \tag{4.66}$$

therefore,

$$\begin{aligned}
\nabla \cdot \mathbf{A}\Psi(\mathbf{r},t) &= \Psi(\mathbf{x},t)\nabla \cdot \mathbf{A} + \mathbf{A}\cdot\nabla\Psi(\mathbf{r},t) \\
&= \mathbf{A}\cdot\nabla\Psi(\mathbf{r},t),
\end{aligned} \tag{4.67}$$

and we back to the Schrödinger's equation with the substitution of Eq. 4.67, and yield,

$$\begin{aligned}
i\hbar \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\frac{\partial f(t)}{\partial t} &= \frac{1}{2m}\left\{p^2 + \left[\frac{e\mathbf{E}}{\omega}\right]^2 \sin^2 \omega t + 2\mathbf{p}\frac{e\mathbf{E}}{\omega}\sin\omega t\right\} \\
&\quad \times \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) f(t) \qquad (4.68) \\
&= \frac{1}{2m}\left(\mathbf{p} + \frac{e\mathbf{E}}{\omega}\sin\omega t\right)^2 \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) f(t). \qquad (4.69)
\end{aligned}$$

Then, by solving the differential equation,

$$\begin{aligned}
\int \frac{\partial f(t)}{f(t)} &= -\frac{i}{\hbar}\left\{\frac{1}{2m}\int\left(\mathbf{p} + \frac{e\mathbf{E}}{\omega}\sin\omega t\right)^2 dt\right\}, \qquad (4.70) \\
\ln f(t) &= -\frac{i}{\hbar}\left\{\frac{1}{2m}\int_0^t\left(\mathbf{p} + \frac{e\mathbf{E}}{\omega}\sin\omega\tau\right)^2 d\tau\right\}, \qquad (4.71)
\end{aligned}$$

obviously we can obtain the solution of the time-dependent function

$$f(t) = \exp\left\{-\frac{i}{\hbar}\left[\frac{1}{2m}\int_0^t\left(\mathbf{p} + \frac{e\mathbf{E}}{\omega}\sin\omega\tau\right)^2 d\tau\right]\right\}. \tag{4.72}$$

Next, we retrieve the Volkov wavefunction by substitute the time-dependent function $f(t)$ into Eq. 4.63,

$$\begin{aligned}
\Psi(\mathbf{r},t) &= \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) f(t) \\
&= \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\exp\left\{-\frac{i}{\hbar}\left[\frac{1}{2m}\int_0^t\left(\mathbf{p} + \frac{e\mathbf{E}}{\omega}\sin\omega\tau\right)^2 d\tau\right]\right\}
\end{aligned} \tag{4.73}$$

Furthermore, we apply the length gauge transformation on Eq. 4.73 and yield

$$\begin{aligned}\Psi_P(\mathbf{r},t) &= \exp\left[-\frac{i}{\hbar}\mathbf{A}(t)\cdot\mathbf{r}\right]\Psi(\mathbf{r},t) &(4.74)\\ &= \exp\frac{i}{\hbar}\left\{\left[\mathbf{p}+\frac{e\mathbf{E}}{\omega}\sin\omega t\right]\cdot\mathbf{r}-\left[\frac{1}{2m}\int_0^t\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}\sin\omega\tau\right)^2 d\tau\right]\right\} &(4.75)\end{aligned}$$

Finally, the Volkov wavefunction as in Eq. 4.54 is obtained.

4.3.2 Keldysh's formalism

As the starting of Keldysh's formalism, firstly, we use the hydrogen s-th bound state at $n=1$, $l=0$ and $m=0$ as the initial state for the tunnelling ionization

$$\begin{aligned}\psi(\mathbf{r}) &\rightarrow \psi_{100}(\mathbf{r},\theta,\phi)\\ &= \left(\frac{1}{\pi a^3}\right)\exp\left(-\frac{\mathbf{r}}{a}\right) &(4.76)\end{aligned}$$

where $a=a_0/Z$

For the transition matrix, we have

$$V_{0s}(\mathbf{p},t)=\int\Psi_p^*(\mathbf{r},t)\,e\mathbf{E}(\mathbf{r},t)\cdot\mathbf{r}\Psi_s(\mathbf{r},t)\,d^3r, \quad (4.77)$$

r-vector take $r(\sin\theta\cos\phi,\sin\theta\sin\phi,\cos\theta)$ and the dot product for the electric field $\mathbf{E}$ and directionality vector $\mathbf{r}$ is

$$\begin{aligned}\mathbf{E}\cdot\mathbf{r} &= E(a_x,a_y,a_z)\cdot r(\sin\theta\cos\phi,\sin\theta\sin\phi,\cos\theta) &(4.78)\\ &= Er[(a_x\cos\phi+a_y\sin\phi)\sin\theta+a_z\cos\theta] &(4.79)\end{aligned}$$

We make an assumption so that the momentum of the photoelectron is parallel to the direction of the electric field, $\mathbf{E}$. Hence,

$$\mathbf{p}\cdot\mathbf{r}=pr[(a_x\cos\phi+a_y\sin\phi)\sin\theta+a_z\cos\theta]$$

Since our case is for linearly polarized laser field, therefore we choose the propagation is along the z-direction. Next, we separate Eq. 4.77 into two parts, the matrix element prefactor (spatial part) and also the action part (time dependent part).

The matrix element is defined as the following expression

$$
\begin{aligned}
V_0\left(\Pi\left(t\right)\right) &= \left(\frac{1}{\pi a^3}\right)^{\frac{1}{2}}\times \\
& \int \exp\left[-\frac{i}{\hbar}\Pi\left(t\right)\cdot\mathbf{r}\right] e\mathbf{E}\cdot\mathbf{r}\exp\left(-\frac{\mathbf{r}}{a}\right)d^3r
\end{aligned}
\tag{4.80}
$$

meanwhile the action part is defined as

$$
\begin{aligned}
e^{iS(\mathbf{p},t)} &= e^{i\Omega(p)t}\times \\
& \exp\left\{-\frac{i}{\hbar}\left[\frac{e\mathbf{p}\cdot\mathbf{E}}{m\omega}\left(\cos\omega t-1\right)+\frac{e^2\mathbf{E}^2}{8m\omega^2}\sin 2\omega t\right]\right\}
\end{aligned}
\tag{4.81}
$$

where

$$
\Omega\left(p\right) = \frac{1}{\hbar}\left(I_0+\frac{p^2}{2m}+\frac{e^2E^2}{4m\omega^2}\right) \tag{4.82}
$$

$$
= \frac{1}{\hbar}\left(I_0+K+U_p\right) \tag{4.83}
$$

Next, we solve for the matrix element prefactor. By using the transformation $u = \sin\omega t$, the result yields

$$
V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right) = -i\frac{2\pi}{\hbar}\sqrt{\frac{1}{\pi a^3}}e16a^5I_0^3\frac{\mathbf{E}\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)}{\left[\frac{\hbar^2}{2ma^2}+\frac{\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)^2}{2m}\right]^3} \tag{4.84}
$$

Meanwhile, for the action part, we set

$$
\widetilde{I}_0 = I_0+U_p \tag{4.85}
$$

hence,

$$
\begin{aligned}
S\left(\mathbf{p},t\right) &= \frac{1}{\hbar}\left[\left(\widetilde{I}_0+\frac{p^2}{2m}\right)t-\frac{e\mathbf{p}\cdot\mathbf{E}}{m\omega}\left(\cos\omega t-1\right)\right. \\
& \left.-\frac{e^2\mathbf{E}^2}{8m\omega^2}\sin 2\omega t\right]
\end{aligned}
\tag{4.86}
$$

We perform the same transformation as in matrix element prefactor Eq. 4.86 can bee expressed as

$$
\begin{aligned}
S(\mathbf{p},u) &= \frac{1}{\hbar\omega}\left[\left(\widetilde{I}_0+\frac{p^2}{2m}\right)\sin^{-1}u\right. \\
&\quad \left.-\frac{e\mathbf{p}\cdot\mathbf{E}}{m\omega}\left(\sqrt{1-u^2}-1\right)-\frac{e^2\mathbf{E}^2}{8m\omega^2}u\sqrt{1-u^2}\right] \\
&= N\sin^{-1}u-a\left(\sqrt{1-u^2}-1\right)-bu\sqrt{1-u^2} \qquad (4.87)
\end{aligned}
$$

where N is the number of photon

$$
\begin{aligned}
N &= \frac{\widetilde{I}_0}{\hbar\omega}+\frac{\chi^2 I_0}{\hbar\omega} \\
&= \frac{\widetilde{I}_0}{\hbar\omega}+2\gamma^2\chi^2 b, \qquad (4.88)
\end{aligned}
$$

with the coefficient a is defined as

$$
\begin{aligned}
a &= \frac{1}{\hbar\omega}\frac{e\mathbf{p}\cdot\mathbf{E}}{m\omega} \\
&= \frac{2I_0}{\gamma\hbar\omega}\xi\chi \\
&= (4b\gamma)\,\xi\chi, \qquad (4.89)
\end{aligned}
$$

and the coefficient b which is

$$
\begin{aligned}
b &= \frac{1}{\hbar\omega}\frac{e^2\mathbf{E}^2}{8m\omega^2} \\
&= \frac{U_p}{\hbar\omega}, \qquad (4.90)
\end{aligned}
$$

and

$$
\xi = \cos\theta. \qquad (4.91)
$$

The momentum depends on n through the following expression

$$
\begin{aligned}
\chi &= \frac{p_n}{\sqrt{2mI_0}} \\
&= \sqrt{\frac{\hbar\omega}{I_0}(n-n_0)}, \qquad (4.92)
\end{aligned}
$$

where $n_0=\frac{I_0+U_p}{\hbar\omega}$ and $p_n=\sqrt{2m\hbar\omega(n-n_0)}$.

For linearly polarized, the ionization rate is for small momentum was first derived by Keldysh in (Keldysh, 1965) . The theory is valid for small momenta such that terms higher than $(\frac{p}{\sqrt{2mI_0}})^2$ are negligible. This restriction also implies a limitation to the laser field E (since $\frac{d\mathbf{p}}{dt} \simeq eE$) and hence the Keldysh parameter γ.

The Keldysh parameter has the alternative statements,

$$\gamma = \sqrt{\frac{E_B}{2U_p}} \tag{4.93}$$

$$= \frac{\omega}{I^{1/2}}\sqrt{2E_B} \tag{4.94}$$

$$= \sqrt{\frac{I_0}{2U_p}}, \tag{4.95}$$

where E_B is the field free binding energy of the electron in the atom or I_0 the ionization potential of the atom.

U_p is the ponderomotive energy (the interaction energy during the transition) of the free electron in the field, and ω is the frequency of the ionization field of intensity I.

We combine the matrix element prefactor and the action and rewrite it in the form

$$L(\mathbf{p}) = \frac{1}{2\pi}\oint V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)\exp\left\{\frac{i}{\hbar\omega}\int_0^u I_0 + \frac{1}{2m}\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)^2\right\}. \tag{4.96}$$

4.3.3 Saddle Point Solution

By defining the transformation

$$u = \sin x, \tag{4.97}$$

$$\frac{du}{dx} = \cos x, \tag{4.98}$$

with $x = \omega t$. Then we can rewrite the $L(\mathbf{p})$ function as

$$L(\mathbf{p}) = \frac{1}{2\pi}V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right) \tag{4.99}$$
$$\times\exp\left\{\int_0^u \frac{1}{\sqrt{1-v^2}}\left[I_0+\frac{1}{2m}\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}v\right)\right]dv\right\}du,$$

with the saddle point u_s which depends on p_s.

Next, we apply Saddle point method on Eq. 4.96,

$$\begin{aligned} \frac{dS(\mathbf{p},t)}{dt} &= \frac{1}{\hbar}\left[I_0+\frac{1}{2m}\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}\sin\omega t\right)^2\right] \\ &= 0. \end{aligned} \tag{4.100}$$

Two saddle point are obtain from Eq. 4.100, which are

$$\begin{aligned} u_\pm &= \frac{1}{4b}\left[-a\pm\sqrt{(a^2-8bN+8b^2)}\right] \\ &= -y\pm\sqrt{y^2+\frac{b-N}{2b}} \\ &\approx \gamma\left[-x\cos\theta\pm i\left(1+x^2\frac{1}{2}\sin^2\theta\right)\right], \end{aligned} \tag{4.101}$$

and the relation of both saddle points are

$$u_- = u_+^*. \tag{4.102}$$

Similarly, we obtain the expression for $x=\omega t$ subsequently from the result of both saddle points,

$$\omega t_+ \simeq i\sinh^{-1}\gamma-\frac{\gamma\cos\theta}{\sqrt{1+\gamma^2}}x+\frac{1}{2}i\gamma\frac{\sin^2\theta+\gamma^2}{\left(\sqrt{1+\gamma^2}\right)^3}x^2, \tag{4.103}$$

$$\omega t_- \simeq \pi+i\sinh^{-1}\gamma+\frac{\gamma\cos\theta}{\sqrt{1+\gamma^2}}x+\frac{1}{2}i\gamma\frac{\sin^2\theta+\gamma^2}{\left(\sqrt{1+\gamma^2}\right)^3}x^2. \tag{4.104}$$

The first term in Eq. 4.103 dominants the imaginary part $i\sinh^{-1}\gamma$ must be positive so that exponential of the function $\exp[iS(t_+)]$ will decay exponentially instead of blowing up. Therefore, we calculate for the following expression

$$\cos\omega t_+ = \sqrt{1+\gamma^2}+i\frac{\gamma^2\cos\theta}{\sqrt{1+\gamma^2}}x+\frac{1}{2}\gamma^2\frac{(1+\gamma^2)\sin^2\theta-\cos^2\theta}{\left(\sqrt{1+\gamma^2}\right)^3}x^2, \tag{4.105}$$

$$\cos\omega t_- = -\sqrt{1+\gamma^2}+i\frac{\gamma^2\cos\theta}{\sqrt{1+\gamma^2}}x-\frac{1}{2}\gamma^2\frac{(1+\gamma^2)\sin^2\theta-\cos^2\theta}{\left(\sqrt{1+\gamma^2}\right)^3}x^2. \tag{4.106}$$

After that, we proceed to the second order of the action part and yield

$$S''(u) = \frac{\frac{e}{m\omega}\mathbf{E}\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)}{\hbar\omega\sqrt{1-u^2}} + \frac{u\left[I+\frac{1}{2m}\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)^2\right]}{\left(1-u^2\right)^{3/2}}. \tag{4.107}$$

Then, we rewrite the expression Eq. 4.107 and get

$$\begin{aligned} S''(u_s) &= \frac{\frac{e}{m\omega}\mathbf{E}\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u_s\right)}{\hbar\omega\sqrt{1-u_s^2}} \\ &= \pm 4\frac{U_p}{\hbar\omega}\frac{i\gamma\sqrt{1+\chi^2 s^2}}{\sqrt{1-u_\pm^2}} \\ &= \pm\frac{2U_p}{\hbar\omega}\frac{u_+ - u_-}{\sqrt{1-u_\pm^2}} \end{aligned} \tag{4.108}$$

In view of the delta function $\delta\left(\hbar A(p) - n\hbar\omega\right)$ where u_s depends on n only and the momenta which is satisfying it are finite. Hence we rewrite it as

$$u_{n\pm} = u_{n\pm}(p_n) \tag{4.109}$$

$$= -\xi\sqrt{\frac{\frac{p_n^2}{2m}}{2U_p}} \pm i\sqrt{\frac{I_0+\frac{p_n^2}{2m}\left(1-\xi^2\right)}{2U_p}}, \tag{4.110}$$

where $\chi = \frac{p_n}{\sqrt{2mI_0}}$ contains all the n dependence. Then the action part with repect of each saddle point becomes

$$S(u_\pm) = \left(\frac{\widetilde{I}_0}{\hbar\omega} + 2\gamma^2 x^2 b\right)\omega t_\pm - \left(4\gamma\chi\xi + \sin\omega t_\pm\right) b\cos\omega t_\pm + 4\gamma\xi x b \tag{4.111}$$

$$= \left(\frac{\widetilde{I}_0}{\hbar\omega} + 2\gamma^2 x^2 b\right)\sin^{-1}\left[\gamma\left(-\xi x \pm i\sqrt{1+x^2 s^2}\right)\right] + 4\gamma\xi x b \tag{4.112}$$

$$-\left(4\xi x - \xi x \pm i\sqrt{1+x^2 s^2}\right) b\sqrt{1-\gamma^2\left(-\xi x \pm i\sqrt{1+x^2 s^2}\right)^2}$$

with

$$\begin{aligned} S(u_+) &= i\left(\frac{I_0+U_p}{\hbar\omega}\sinh^{-1}\gamma - \frac{U_p}{\hbar\omega}\gamma\sqrt{1+\gamma^2}\right) \\ &\quad + 4\gamma\cos\theta\frac{U_p}{\hbar\omega}\left(1-\sqrt{1+\gamma^2}x\right) + i2\gamma^2\frac{U_p}{\hbar\omega}\left(\sinh^{-1}\gamma - \frac{\gamma\cos^2\theta}{\sqrt{1+\gamma^2}}\right)x^2 \end{aligned} \tag{4.113}$$

$$= -A + (B-C)x + iDx^2 \tag{4.114}$$

and

$$
\begin{aligned}
S(u_-) &= i\left(\frac{I_0+U_p}{\hbar\omega}\sinh^{-1}\gamma-\frac{U_p}{\hbar\omega}\gamma\sqrt{1+\gamma^2}\right) && (4.115)\\
&\quad +4\gamma\cos\theta\frac{U_p}{\hbar\omega}\left(1+\sqrt{1+\gamma^2}x\right)+i2\gamma^2\frac{U_p}{\hbar\omega}\left(\sinh^{-1}\gamma-\frac{\gamma\cos^2\theta}{\sqrt{1+\gamma^2}}\right)x^2\\
&\quad +\pi\left(\frac{I_0+U_p}{\hbar\omega}+2\gamma^2\frac{U_p}{\hbar\omega}x^2\right)\\
&= -A+(B+C)x+iDx^2+\pi F && (4.116)
\end{aligned}
$$

Next, by using the following expression

$$
\begin{aligned}
S''(u) &= \frac{dS(\mathbf{p},u)}{du} && (4.117)\\
&= \frac{\frac{e}{m\omega}E\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)}{\hbar\omega\sqrt{1-u^2}}, && (4.118)
\end{aligned}
$$

and

$$
I_0=\frac{\hbar^2}{2ma^2}, \tag{4.119}
$$

then the $L(\mathbf{p})$ is rewritten as

$$
\begin{aligned}
L(\mathbf{p}) &= \frac{16ieI_0^3\sqrt{\pi a^7}}{\hbar^2\omega}\sum_s\frac{\mathbf{E}\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u_s\right)}{(\hbar\omega)^2S''(u_s)^2\left(\sqrt{1-u_s^2}\right)^3}e^{iS(\mathbf{p},u_s)} && (4.120)\\
&= \frac{4\hbar\omega I_0\sqrt{\pi a}}{eE}\sum_{s=\pm}\frac{e^{iS(\mathbf{p},u_s)}}{\cos\Theta^s\cos\omega t_s} && (4.121)
\end{aligned}
$$

As a result, we have

$$
|L(\mathbf{p}_n)|^2=\left(\frac{4\hbar\omega I_0}{eE}\right)^2\pi a\left|\frac{e^{iS(\mathbf{p},u_+)}}{\cos\Theta^+\cos\omega t_+}+\frac{e^{iS(\mathbf{p},u_-)}}{\cos\Theta^-\cos\omega t_-}\right|^2, \tag{4.122}
$$

where

$$
\cos\Theta^s=\pm\sqrt{1+\frac{p^2\sin^2\theta}{2mI_0}}. \tag{4.123}
$$

4.4 Rate for small momentum

In Keldysh's work in 1965, he made an assumption so that in the photoionization system, the momentum of the photoelectron is very small. Following by the Eq. 4.122, if we only consider the first order of the photoelectron momentum after expanding, then we have

$$L(\mathbf{p}) = \frac{4\hbar\omega I_0\sqrt{\pi a}}{eE}\frac{e^{iS(\mathbf{p},u_+)}+e^{iS(\mathbf{p},u_-)}}{\sqrt{1+\gamma^2}}, \tag{4.124}$$

and subsequently

$$|L(\mathbf{p}_n)|^2 = \left(\frac{4\hbar\omega I_0}{eE}\right)^2\frac{\pi a}{1+\gamma^2}\left|e^{iS(\mathbf{p},u_+)}+e^{iS(\mathbf{p},u_-)}\right|^2. \tag{4.125}$$

Therefore, the integral of Eq. 4.125 yields

$$\begin{aligned}\int_0^\pi |L(\mathbf{p}_n)|^2\sin\theta d\theta &= \left(\frac{4\hbar\omega I_0}{eE}\right)^2\frac{2\pi a}{1+\gamma^2}\exp\left(-2n_0\sinh^{-1}\gamma+2b\gamma\sqrt{1+\gamma^2}\right) \quad (4.126)\\ &\times\exp\left[-(n-n_0)\left(\sinh^{-1}\gamma-\frac{\gamma\cos^2\theta}{\sqrt{1+\gamma^2}}\right)\right]\\ &= \left(\frac{4\hbar\omega I_0}{eE}\right)^2\frac{4\pi a}{1+\gamma^2}\left(\frac{\sqrt{1+\gamma^2}}{2\gamma(n-n_0)}\right)^{\frac{1}{2}} \quad (4.127)\\ &\times\exp\left\{2(n-n_0)\left[\frac{\gamma}{\sqrt{1+\gamma^2}}-\sinh^{-1}\gamma+2n_0\left(\frac{\gamma\sqrt{1+\gamma^2}}{2\gamma^2+1}-\sinh^{-1}\gamma\right)\right]\right\}\end{aligned}$$

Consequently, the celebrated formula for ionization rate written as

$$\begin{aligned}w &= 8\omega\sqrt{\frac{2I_0}{\hbar\omega}}\xi^{3/2}\exp[2n_0(\frac{\gamma\sqrt{1+\gamma^2}}{2\gamma^2+1}-\sinh^{-1}\gamma)]\\ &\times\sum_{n=n_0}^{\infty}\exp\left[2\Delta n(\xi-\sinh^{-1}\gamma)\right]\mathscr{D}(\sqrt{2\xi\Delta n}) \quad (4.128)\end{aligned}$$

where $\Delta n = n-n_0$, $\xi = \frac{\gamma}{\sqrt{1+\gamma^2}}$, $I_0 = \frac{\hbar^2}{2ma^2}$ is the ionization energy with the Bohr radius a, $U_p = \frac{e^2E^2}{4m\omega^2}$ is the ponderomotive energy, $n_0(E,\omega) = \frac{I_0+U_p}{\hbar\omega}$ with the Dawson integral

$$\begin{aligned}\mathscr{D}(y) &= \int_0^y \exp(z^2-y^2)dz \quad (4.129)\\ y^2 &= 2\xi\Delta n \quad (4.130)\end{aligned}$$

Eq. 4.128 is two times larger than that in Eq. 4.177 because the two poles are included. We will discuss this matter further in the following section.

4.4.1 Exact rate for arbitrary momentum

In this case, we have extended the limit of Keldysh's model by taking consideration of the higher order term (Long & Liu, 2011) of the photoelectron momentum. In other word, our exact model can adopt arbitrary value for the photoelectron momentum. Therefore, we continue from the $L(\mathbf{p},t)$ function which is

$$\begin{aligned} L(\mathbf{p},t) &= V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)\exp\left\{\frac{i}{\hbar\omega}\int_0^u I_0+\frac{1}{2m}\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)^2\right\} && (4.131)\\ &= V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)e^{iS(\mathbf{p},t)}, && (4.132)\end{aligned}$$

where $S(\mathbf{p},t)$ is the action part

$$S(\mathbf{p},t)=\Omega(p)\,t-\frac{1}{\hbar\omega}\left[\frac{e\mathbf{p}\cdot\mathbf{E}}{m\omega}(\cos\omega t-1)+\frac{e^2\mathbf{E}^2}{8m\omega^2}\sin 2\omega t\right], \tag{4.133}$$

with the conservation of energy term

$$\begin{aligned} \Omega(p) &= \frac{1}{\hbar}\left(I_0+\frac{p^2}{2m}+\frac{e^2E^2}{4m\omega^2}\right) && (4.134)\\ &= \frac{1}{\hbar}\left(I_0+K+U_p\right). && (4.135)\end{aligned}$$

Next, we perform the Fourier series on $L(\mathbf{p},t)$ function by expanding on the t term and yield

$$\begin{aligned} L(\mathbf{p},t) &= \sum_{n=-\infty}^{\infty} L_n(\mathbf{p})\exp\left[i(\Omega-n\omega)t\right] && (4.136)\\ &= V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)e^{iS(\mathbf{p},t)}, && (4.137)\end{aligned}$$

with

$$V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)=-i\frac{2\pi}{\hbar}\sqrt{\frac{1}{\pi a^3}}e16a^5I_0^3\frac{\mathbf{E}\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)}{\left[\frac{\hbar^2}{2ma^2}+\frac{\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)^2}{2m}\right]^3}. \tag{4.138}$$

Hence, we find the new expression for $L_n(\mathbf{p})$ is a n-dependent function where the n is the number of photon which is proportional to the intense laser source

$$L_n(\mathbf{p}) = \frac{1}{2\pi}\int_{-T/2}^{T/2} L(\mathbf{p},t)\exp\left[-i(\Omega - n\omega)t\right]\omega dt \tag{4.139}$$

$$= \frac{1}{2\pi}\int_{-T/2}^{T/2} V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)e^{iS(\mathbf{p},t)}\exp\left[-i(\Omega - n\omega)t\right]\omega dt \tag{4.140}$$

In order to expand the new $L_n(\mathbf{p})$ function with respect of the number of photon, n, note that

$$L_{n\pm1}(\mathbf{p}) = \frac{1}{2\pi}\int_{-T/2}^{T/2} V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)e^{\pm i\omega t} \tag{4.141}$$
$$\times\exp\left\{\frac{1}{\hbar}\left[n\hbar\omega - \frac{e\mathbf{p}\cdot\mathbf{E}}{m\omega}(\cos\omega t - 1) + \frac{e^2\mathbf{E}^2}{8m\omega^2}\sin 2\omega t\right]\right\}\omega dt,$$

and

$$L_{n+1}(\mathbf{p}) + L_{n-1}(\mathbf{p}) = \frac{1}{2\pi}\int_{-T/2}^{T/2}(2\cos\omega t)V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)e^{\pm i\omega t} \tag{4.142}$$
$$\times\exp\left\{\frac{1}{\hbar}\left[n\hbar\omega - \frac{e\mathbf{p}\cdot\mathbf{E}}{m\omega}(\cos\omega t - 1) + \frac{e^2\mathbf{E}^2}{8m\omega^2}\sin 2\omega t\right]\right\}\omega dt$$
$$= \frac{1}{2\pi}\int_{-T/2}^{T/2}(2\cos\omega t)L(\mathbf{p},t)\exp\left[i(n\omega - \Omega)t\right]\omega dt. \tag{4.143}$$

Then, the inversion of the Eq. 4.143 gives us

$$L(\mathbf{p},t)\cos\omega t = \frac{1}{2}\sum_{n=-\infty}^{\infty}\Lambda_n(\mathbf{p})\exp\left[i(\Omega - n\omega)t\right] \tag{4.144}$$

$$= \frac{1}{2}\sum_{n=-\infty}^{\infty}L_n(\mathbf{p})\left[e^{i[(\Omega-(n+1)\omega)]t} + e^{i[(\Omega-(n-1)\omega)]t}\right], \tag{4.145}$$

where

$$\Lambda_n(\mathbf{p}) = L_{n+1}(\mathbf{p}) + L_{n-1}(\mathbf{p}). \tag{4.146}$$

4.4.1 (a) Residue Theorem

From Eq. 4.138, obviously we can see that $V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)$ has singularity, hence we might rewrite the $L(\mathbf{p})$ function as the following expression

$$L(\mathbf{p}) = -16i\frac{eI_0^3}{\hbar}\sqrt{\frac{a^7}{\pi}}\frac{\mathbf{E}\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)}{\left[\hbar\omega S'(u)\sqrt{1-u^2}\right]^3}e^{iS(\mathbf{p},u)}du. \tag{4.147}$$

Then, we further expand on $S'(u)$ and yield

$$L(\mathbf{p}) = -\frac{16i\frac{eI_0^3}{\hbar}\sqrt{\frac{a^7}{\pi}}}{\left[\hbar\omega S''(u_s)\right]^3}\frac{f(u)}{(u-u_s)^3}du, \tag{4.148}$$

with

$$f(u) = \frac{\mathbf{E}\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)e^{iS(\mathbf{p},u)}}{\left(\sqrt{1-u^2}\right)^3}. \tag{4.149}$$

Next, we apply residue theorem to find the singularities. For simplification purpose, let us define $X = e^{iS(\mathbf{p},u)}$, then the first order differentiation on function $f(u)$ with respect of u is

$$\frac{df(u)}{du} = \frac{\mathbf{E}\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)}{\left(\sqrt{1-u^2}\right)^3}\frac{dX}{du} + XE\frac{Ee+2eEu^2+3up\omega}{\omega\left(\sqrt{1-u^2}\right)^{\frac{5}{2}}}, \tag{4.150}$$

and the second order differentiation yields

$$\frac{d^2f(u)}{du^2} = J+K+L, \tag{4.151}$$

with

$$\begin{aligned} J &= \frac{\mathbf{E}\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)}{\left(\sqrt{1-u^2}\right)^3}\frac{d^2X}{du^2}, && (4.152)\\ K &= 2\frac{dX}{du}E\frac{Ee+2eEu^2+3up\omega}{\omega\left(\sqrt{1-u^2}\right)^{\frac{5}{2}}}, && (4.153)\\ L &= 3XE\frac{\left(p+\frac{e\mathbf{E}}{\omega}u\right)\left(3+2u^2\right)-\left(1-u^2\right)2p}{\left(1-u^2\right)^3\left(\sqrt{1-u^2}\right)}. && (4.154) \end{aligned}$$

As we look at the Eq. 4.151, we can see that

$$\begin{aligned} \frac{dX}{du} &= iS'X && (4.155)\\ &= 0, && (4.156) \end{aligned}$$

hence, Eq. 4.151 becomes

$$\frac{d^2}{du^2}f(u) = iS''e^{iS(\mathbf{p},u)}\frac{\mathbf{E}\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u\right)}{\left(\sqrt{1-u^2}\right)^3}. \tag{4.157}$$

Then, we obtain the final expression for $L(\mathbf{p})$ function which is

$$L(\mathbf{p}) = \frac{16i\frac{eI_0^3}{\hbar}\sqrt{\pi a^7}}{(\hbar\omega)^3}\sum_s \frac{\mathbf{E}\cdot\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}u_s\right)}{S''(u_s)^2\left(\sqrt{1-u_s^2}\right)^3}e^{iS(\mathbf{p},u_s)}. \tag{4.158}$$

4.4.1 (b) *Transition Probability and Ionization Rate*

Before we further proceed, let us define a new term $c_b(\mathbf{p},t)$ which is known as the transition coefficient. It describes the process where the electron is transited from the hydrogen ground state to the continuum state in the period of time t. We recall the transition matrix $V_{0s}(\mathbf{p},t)$ as in Eq. 4.77 and yield

$$\begin{aligned} c_b(\mathbf{p},t) &= \frac{1}{i\hbar}\int_{-\infty}^{t} V_{0s}(\mathbf{p},t')\,dt' && (4.159)\\ &= \frac{1}{i\hbar}\int_{-\infty}^{t} L(\mathbf{p},t')\cos\omega t' dt' && (4.160)\\ &= \frac{1}{i\hbar}\int_{-\infty}^{t} V_0\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}\sin\omega t'\right)\cos\omega t' && (4.161)\\ &\quad\times\exp\left\{\frac{i}{\hbar}\int_0^{t'}\left[I_0+\frac{1}{2m}\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}\sin\omega\tau\right)^2 d\tau\right]\right\}dt' \end{aligned}$$

Next, we expand on the coefficient and get

$$\begin{aligned} c_b(\mathbf{p},t) &= \frac{1}{i\hbar}\int_{-\infty}^{t}\left\{\int\exp\left[-\frac{i}{\hbar}\Pi(t')\cdot\mathbf{r}\right]e\mathbf{E}\cdot\mathbf{r}\psi_s d^3r\right\} && (4.162)\\ &\quad\times\cos\omega t'\exp\left\{\frac{i}{\hbar}\int_0^{t'}\left[I_0+\frac{1}{2m}\left(\mathbf{p}+\frac{e\mathbf{E}}{\omega}\sin\omega\tau\right)^2 d\tau\right]\right\}dt' \end{aligned}$$

Then, we compute for the probability of the transition process through the following relation

$$\begin{aligned} P &= |c_b(\mathbf{p},t)|^2 && (4.163)\\ &= \frac{1}{\hbar^2}\int_{-\infty}^{t} V_{0s}(\mathbf{p},t')\,dt'\int_{-\infty}^{t} V_{0s}(\mathbf{p},t'')\,t'' && (4.164) \end{aligned}$$

Subsequently, the ionization rate is obtained via applying differentiation on the probability with respect of the transition time

$$
\begin{aligned}
w &= \frac{dP}{dt} && (4.165)\\
&= \int_{-\infty}^{\infty}\frac{d^3p}{(2\pi\hbar)^3}\left\{\frac{dc_b^*(\mathbf{p},t)}{dt}c_b(\mathbf{p},t)+c_b^*(\mathbf{p},t)\frac{dc_b(\mathbf{p},t)}{dt}\right\} && (4.166)\\
&= \frac{1}{\hbar^2}\int_{-\infty}^{\infty}\frac{d^3p}{(2\pi\hbar)^3}\left[\int_{-\infty}^{t}L^*(t)L(t')\cos\omega t\cos\omega t'dt'\right. && (4.167)\\
&\quad\left.+\int_{-\infty}^{t}L^*(t')L(t)\cos\omega t\cos\omega t'dt'\right]
\end{aligned}
$$

By expanding the $L(t)$ function, the ionization rate becomes

$$
\begin{aligned}
w &= 2Re\frac{1}{\hbar^2}\int_{-\infty}^{\infty}\frac{d^3p}{(2\pi\hbar)^3}\int_{-\infty}^{t}V_0^*(\Pi(t))V_0(\Pi(t')) && (4.168)\\
&\quad\times\exp\left\{\frac{i}{\hbar}\int_t^{t'}\left[I_0+\frac{1}{2m}\Pi(\tau)^2\right]d\tau\right\}\cos\omega t\cos\omega t'dt',
\end{aligned}
$$

and we may write Eq. 4.168 as

$$
\begin{aligned}
w &= \int_{-\infty}^{\infty}\frac{d^3p}{(2\pi\hbar)^3}\frac{1}{4\hbar^2}\sum_{n=-\infty}^{\infty}|\Lambda_n(\mathbf{p})|^2 2\pi\delta(A-n\omega) && (4.169)\\
&= \frac{2\pi}{\hbar}\int_{-\infty}^{\infty}\frac{d^3p}{(2\pi\hbar)^3}\sum_{n=-\infty}^{\infty}\frac{1}{4}|L_{n+1}(\mathbf{p})+L_{n-1}(\mathbf{p})|^2\delta(\hbar A-n\hbar\omega). && (4.170)
\end{aligned}
$$

Let us derive an identity for the function $\Lambda_n(\mathbf{p})$ which is

$$
\begin{aligned}
\frac{1}{2}\Lambda_n(\mathbf{p}) &= \frac{1}{2}[L_{n+1}(\mathbf{p})+L_{n-1}(\mathbf{p})] && (4.171)\\
&= \frac{1}{2\pi}V_0(\Pi(u))e^{iS(\mathbf{p},u)}e^{i(n\omega-\Omega)\frac{1}{\omega}\sin^{-1}u}du. && (4.172)
\end{aligned}
$$

In the aftermath of Eq. 4.172, we have

$$
\begin{aligned}
w &= \frac{2\pi}{\hbar}\int_{-\infty}^{\infty}\sum_{n=-\infty}^{\infty}|L(\mathbf{p})|^2\delta(\hbar\Omega(p)-n\hbar\omega)\frac{d^3p}{(2\pi\hbar)^3} && (4.173)\\
&= \frac{2\pi}{\hbar}m\int_0^{\infty}\int_0^{\pi}\sum_{n=-\infty}^{\infty}\left|\frac{1}{2}\Lambda_n(\mathbf{p})\right|^2\delta(\hbar\Omega(p)-n\hbar\omega)pdE\frac{2\pi\sin\theta d\theta}{(2\pi\hbar)^3}. && (4.174)
\end{aligned}
$$

Next, integration over the energy and the delta function yields

$$
\begin{aligned}
w &= \frac{2\pi}{\hbar} m \int_0^{\pi} \sum_{n=-\infty}^{\infty} |L(\mathbf{p}_n)|^2 p_n \frac{2\pi \sin\theta d\theta}{(2\pi\hbar)^3} && (4.175)\\
&= \frac{m}{2\pi\hbar^4} \int_0^{\pi} \sum_{n=-\infty}^{\infty} |L(\mathbf{p}_n)|^2 p_n \sin\theta d\theta && (4.176)
\end{aligned}
$$

As a result, we have extended the Keldysh's theory to arbitrary momenta, giving the exact result that is semi-analytical,

$$
w = \frac{m}{(2\pi\hbar^2)^2} 2\pi \int_0^{\pi} \sum_{n=n_0}^{\infty} |L(\mathbf{p}_n)|^2 p_n \sin\Theta d\Theta \tag{4.177}
$$

where

$$
|L(\mathbf{p}_n)|^2 = (\frac{4\hbar\omega I_0}{eE})^2 \pi a \left| \frac{e^{iS(\mathbf{p}_n,u_+)}}{\eta_+ \cos\omega t_+} + \frac{e^{iS(\mathbf{p}_n,u_-)}}{\eta_- \cos\omega t_-} \right|^2 \tag{4.178}
$$

with double saddle points

$$
\begin{aligned}
\eta_\pm &= \pm\sqrt{1+\chi^2 \sin^2\Theta} + i\frac{u_\pm^3}{\gamma} && (4.179)\\
u_\pm &= -\gamma\chi a_z \pm \gamma\sqrt{(\chi a_z)^2 - (1+\chi^2)} && (4.180)\\
\omega t_+ &= \sin^{-1} u_+, && (4.181)\\
\omega t_- &= \pi - \sin^{-1} u_- && (4.182)
\end{aligned}
$$

and $a_z = \cos\Theta$.

4.5 Elliptical Polarized

For elliptical polarized light $E = E(\alpha\cos\omega t, \beta\sin\omega t, 0) = \frac{1}{2}E[(\hat{x}\alpha + i\hat{y}\beta)e^{-iwt} + c.c.]$ where α and β determine the ellipticity, the integration over Φ should be included

$$
w = \frac{m}{(2\pi\hbar^2)^2} \int_0^{2\pi} \int_0^{\pi} \sum_{n=n_0'}^{\infty} |L(\mathbf{p}_n)|^2 p_n \sin\Theta d\Theta d\Phi, \tag{4.183}
$$

where

$$
\begin{aligned}
L(\mathbf{p}_n) &= \frac{1}{2\pi} \oint \frac{V_0(\Pi_n(u))}{\sqrt{1-u^2}} e^{iS(\mathbf{p}_n,u)} du \\
&= \frac{1}{2\pi} \int_{-\pi}^{\pi} V_0(\Pi(x)) \exp iS(\mathbf{p}_n,x) dx. && (4.184)
\end{aligned}
$$

Then, the action part for this case would be

$$
\begin{aligned}
S(\mathbf{p},x) &= nx + \frac{eE}{\hbar m\omega^2}\left[\alpha p_x(1-\cos x) - \beta p_y \sin x\right] \\
&\quad -\frac{U_p}{\hbar\omega}\left[\frac{\left(\alpha^2-\beta^2\right)\sin 2x}{2}\right]
\end{aligned}
\tag{4.185}
$$

Here, $n_0' = \frac{I_0+U_p(\alpha^2+\beta^2)}{\hbar\omega}$ and $p_n = \sqrt{2m\hbar\omega(n-n_0')}$ where $u = \sin x$ and $B = \frac{U_p}{\hbar\omega}$. The variable momentum components $p = p(\sin\Theta\cos\Phi, \sin\Theta\sin\Phi, \cos\Theta)$. The subsequent electron momentum of the electron might follow the field predominantly confined in the x-y plane, with $p(a_x, a_y, a_z)$ that can be found from $\frac{d\mathbf{p}}{dt} = e\left(\mathbf{E} + \frac{1}{m}\mathbf{p}\times\mathbf{B}\right)$ which is approximately $\frac{d\mathbf{p}}{dt} \approx eE$, giving $a_z = 0$, and finite transverse components $\frac{a_y}{a_x} \approx \frac{\beta}{\alpha}\frac{u}{\sqrt{1-u^2}}$ that corresponds to setting $\Theta = \pi/2$ in the unit vector of p, i.e. $a_x = \frac{1}{\sqrt{1+(\frac{\beta u}{\alpha v})^2}}, a_y = \frac{1}{\sqrt{1+(\frac{\alpha v}{\beta u})^2}}$.

The transition matrix element between the bound state and the Volkov state is

$$
V_0(t) = e\int \psi_s(\mathbf{r}) E r \sin\theta C(t,\phi) e^{-i\Psi} r^2 dr \sin\theta d\theta d\phi \tag{4.186}
$$

where

$$
\Psi(\theta,\phi) = \frac{1}{\hbar}\left[Qr\sin\theta + Pr\cos\theta\right] \tag{4.187}
$$

$$
C(t,\phi) = M\cos\phi + N\sin\phi, \tag{4.188}
$$

$$
M = \alpha\cos\omega t, N = \beta\sin\omega t \tag{4.189}
$$

$$
P = p_z \tag{4.190}
$$

$$
\begin{aligned}
Q(t,\phi) &= \left(p_x + \frac{eE}{\omega}\alpha\sin\omega t\right)\cos\phi \\
&\quad + \left(p_y - \frac{eE}{\omega}\beta\cos\omega t\right)\sin\phi
\end{aligned}
\tag{4.191}
$$

For hydrogenic atom, $\psi_s(r) = R(r)Y(\theta,\phi) = R(r)\Theta(\theta)\frac{1}{\sqrt{2\pi}}e^{im\phi}$. Assuming the atom in 1s state, $\psi_s(r) = \sqrt{\frac{1}{\pi a^3}}e^{-r/a}$ $\left(R(r) = \sqrt{\frac{4}{a^3}}e^{-r/a}\right)$ we may perform the r integration and obtain a semi-analytical expression

$$
V_0(\Pi(t)) = \sqrt{\frac{1}{\pi a^3}} eE6a^4 \int \{M\cos\phi + N\sin\phi\} G(\phi) d\phi \tag{4.192}
$$

with

$$
G(\phi) = \int \frac{\sin^2\theta d\theta}{\left[1 + i\frac{a}{\hbar}\left(Q\sin\theta + P\cos\theta\right)\right]^4} \tag{4.193}
$$

$$
\begin{aligned}
&= \frac{26Q' - \frac{4Q'^3}{P'^2-1} + 3\pi\left[\frac{4Q'^2}{(P'-1)} - (1+P')\right]\frac{(P'-1)}{i\sqrt{A}}}{6A^3} \\
&\quad + \frac{\left(-1+P'^2-4Q'^2\right)\tanh^{-1}\left(\frac{Q'}{\sqrt{A}}\right)}{A^{\frac{7}{2}}}
\end{aligned}
\tag{4.194}
$$

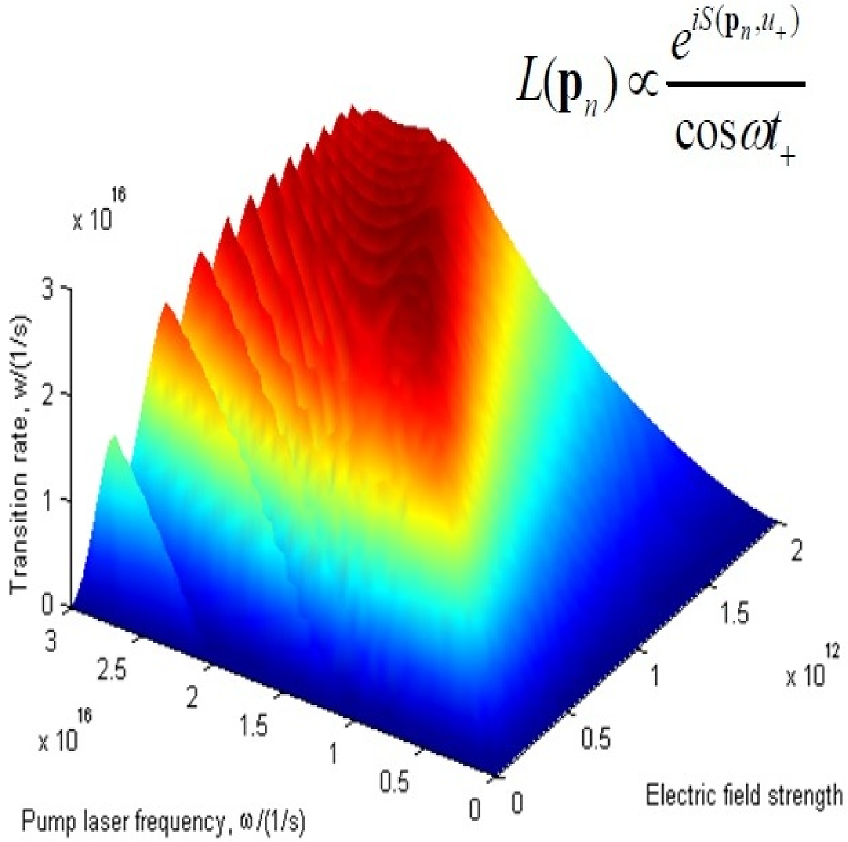


Figure 4.1: Ionization rate versus frequency, ω and electric field, E for positive pole.

where

$$A = P'^2 + Q'^2 - 1 \tag{4.195}$$

$$P' = \frac{ia}{\hbar}P, Q' = \frac{ia}{\hbar}Q \tag{4.196}$$

The above expressions have been computed numerically to obtained results without using the saddle point method from

$$S' = \frac{\Omega}{\omega B} - w + 2wu_s^2 + 4\gamma\chi\{\alpha a_x u_s - \beta a_y\sqrt{1-u_s^2}\} = 0 \tag{4.197}$$

which gives four saddle points u_s with the corresponding derivatives

$$S''(u_s) = \frac{\sqrt{\frac{4U_p}{m}}(\alpha p_x + \beta p_y \frac{u_s}{\sqrt{1-u_s^2}}) + 4U_p u_s(\alpha^2 - \beta^2)}{\hbar\omega\sqrt{1-u_s^2}} \tag{4.198}$$

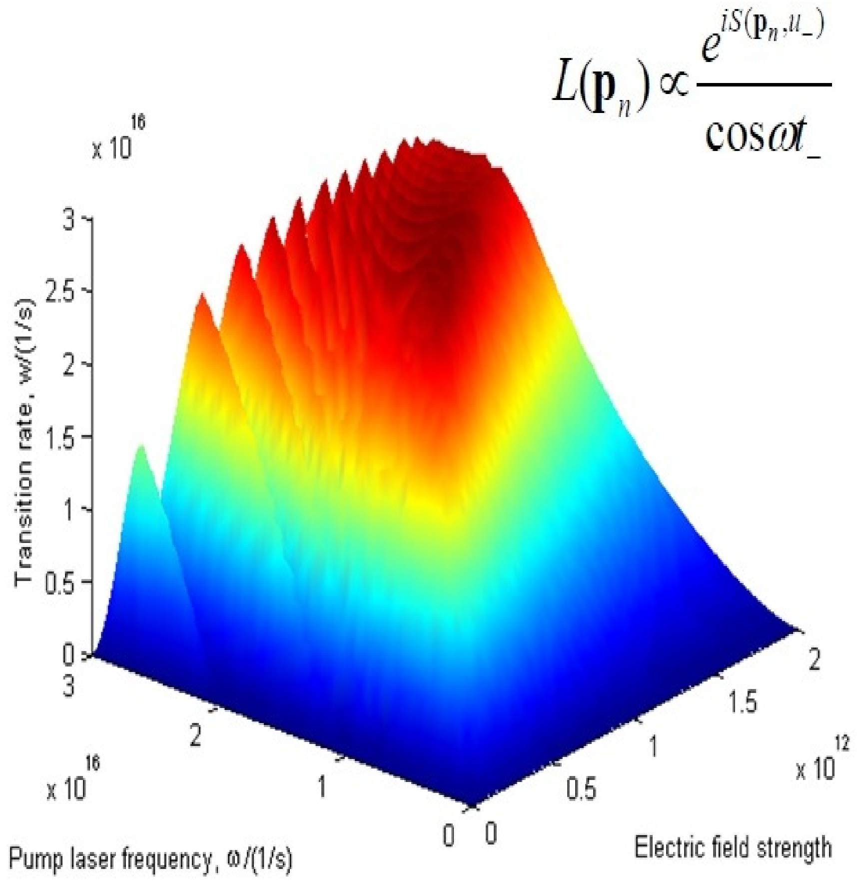


Figure 4.2: Ionization rate versus frequency, ω and electric field, E for negative pole.

4.6 Discussions

In Fig. 4.1, we can see the rate of tunnelling ionization due to the positive pole during the saddle point calculation as in Eqn. 4.128. It is clearly showed that the rate increases with the frequency and electric field strength. We observe the oscillations at the frequency around 10^{16} s^{-1} with the change of electric field. Meanwhile, Fig. 4.2 has shown the rate of tunnelling ionization due to the negative pole. However, the oscillations pattern in Fig. 4.2 is almost having the same shape as in Fig. 4.1. In Fig. 4.3, the exact rate of the tunnelling ionization is computed by taking account of the two poles was shown as the function of frequency and electric field strength. Interesting feature was found due to the interference of the two terms $\frac{e^{iS(\mathbf{p}_n, u_+)}}{\cos \omega t_+}$ and $\frac{e^{iS(\mathbf{p}_n, u_-)}}{\cos \omega t_-}$ in Eq. 4.128 associated with the two saddle points $u_{\pm}$. The inteference in Fig. 4.3 has a increment of a ratio $2\sqrt{\pi/2}$ which is obviously as a result of Eq. 4.177. These multiple poles mainly contribute after taking the consideration of higher order term of momentum in our calculation meanwhile in Keldysh's original work, small momentum approximation was taken for the elimination of higher order momentum term in order to simplify the calculation. In our understand-

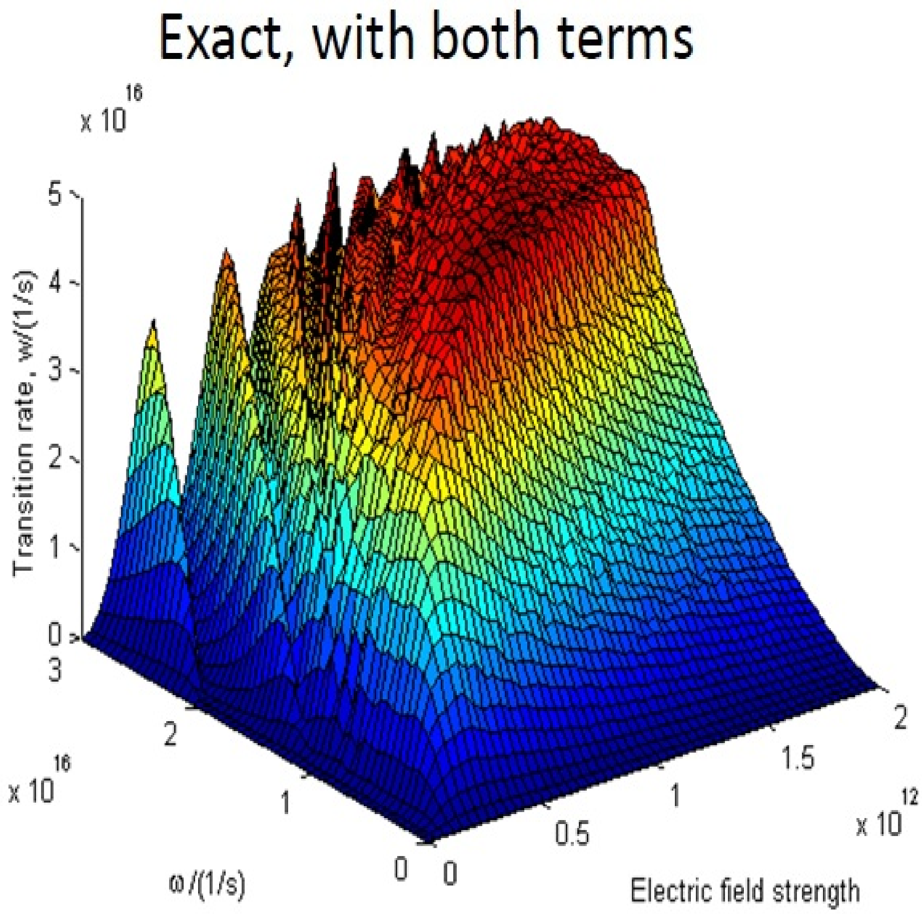
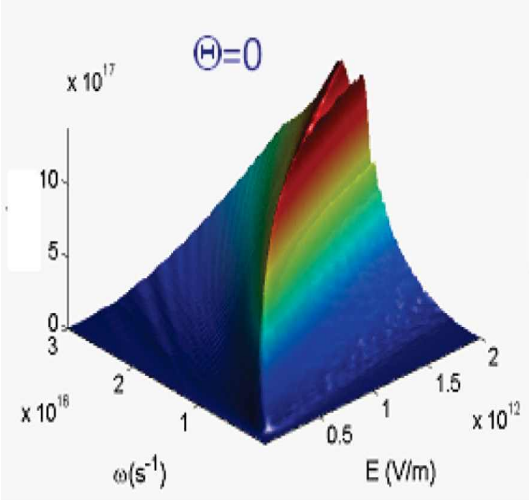


Figure 4.3: Ionization rate versus frequency, ω and electric field, E for both poles included.

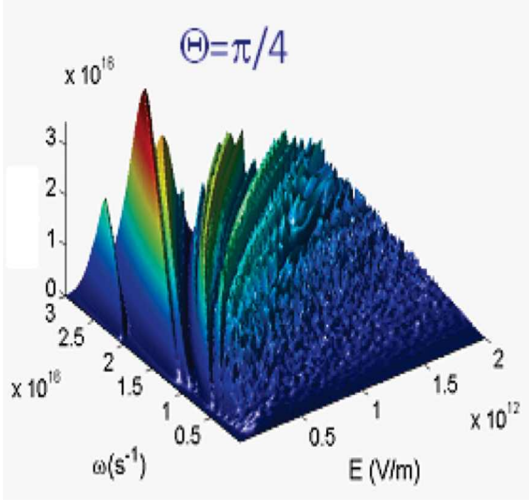
ing, for the first time this interference pattern predominantly appears in the tunnelling ionization rate of photoelectron. Therefore, it would be interesting and also challenging for the further experimental verification. Consequently, our result can take arbitrary value of momentum into account to produce a more accurate photoionization rate.

For linear polarized laser field, we can see that the differential photoionization rate $dw/d\Omega$ via different detection angle Θ as shown in Fig. 4.4. The ionization rate is the maximum as the output source facing the detector which is $\Theta = 0$. As the photoelectron beam is away from the detector as in Fig. 4.4b and Fig. 4.4c, the ionization rate magnitude is decreasing. In the direction of $\Theta = \pi/2$ which mean the direction is perpendicular with the detector, the ionization rate of the photoelectron is still detectable, but in a very low magnitude which is mainly focusing on the regime of high frequency and low electric field strength.

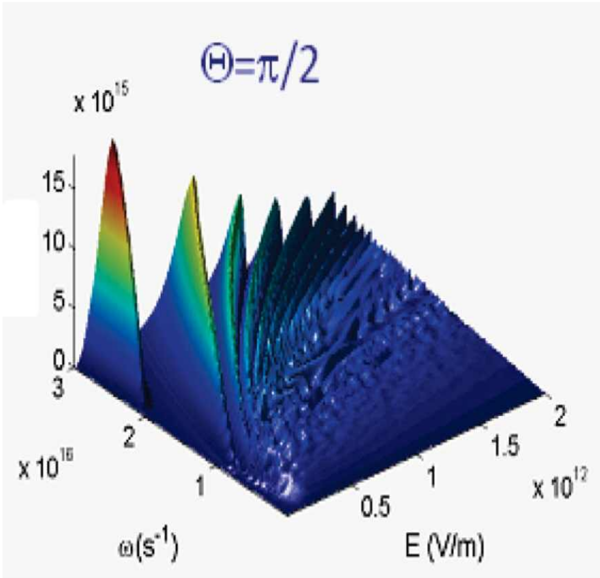
Meanwhile, for circular polarized laser, the differential photoionization rate $dw/d\Omega$



(a) Detection direction $\Theta = 0$.



(b) Detection direction $\Theta = \pi/4$.

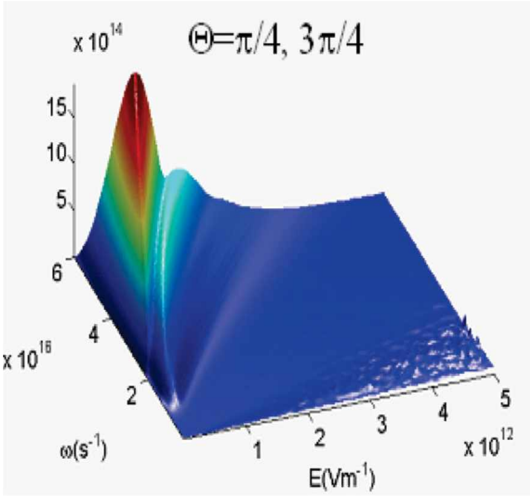


(c) Detection direction $\Theta = \pi/2$.

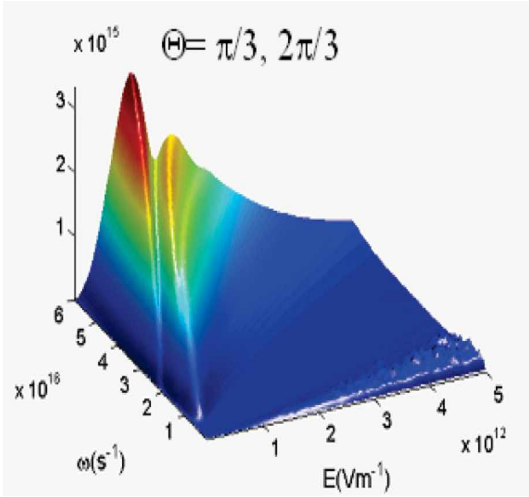
Figure 4.4: Differential ionization rate $dw/d\Omega$ for linear polarized laser field, at different detection angle Θ.

in Fig. 4.5 has the same order of magnitude as the linear polarization. However, the oscillations are more rapid due and this can be traced back to the fourth order root in the saddle point formula Eq. 4.197. The spectra varies little with direction, and quite isotropic.

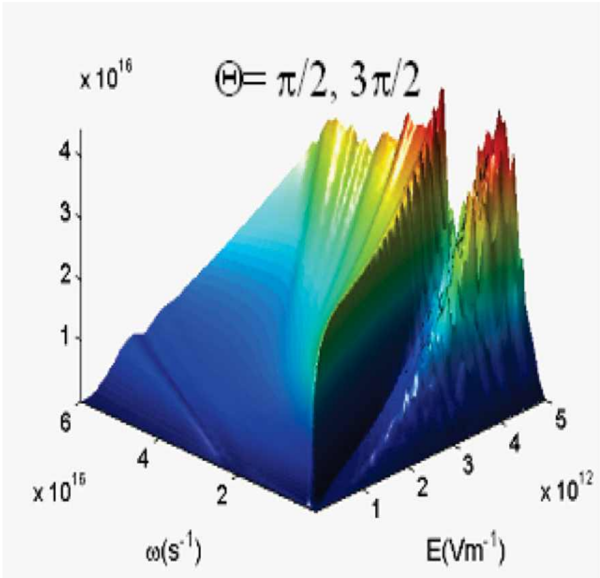
The ellipticity has significant effect on the differential ionization rate spectra and the intensity dependence as in Fig. 4.6. There is a clear minimum threshold frequency of the laser required for photoionization for each value of electric field. The electron is ejected mainly in the x-y plane (when $\Theta = \pi/2$), as expected. The rate along x-direction ($\Phi = 0$) is larger than y-direction, in agreement with recent analysis (Barth & Smirnova, 2011).



(a) Detection direction $\Theta = \pi/4$ and $3\pi/4$.



(b) Detection direction $\Theta = \pi/3$ and $2\pi/3$.

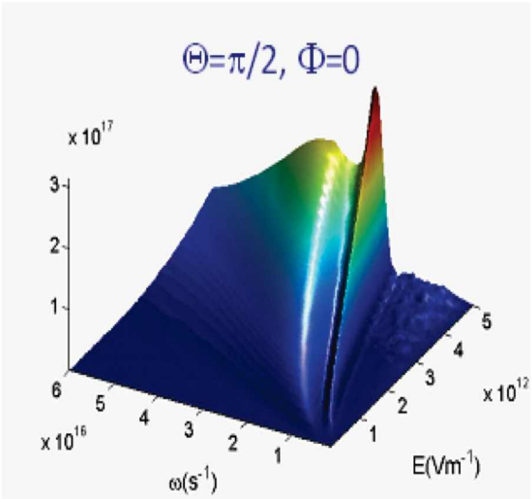


(c) Detection direction $\Theta = \pi/2$ and $3\pi/2$.

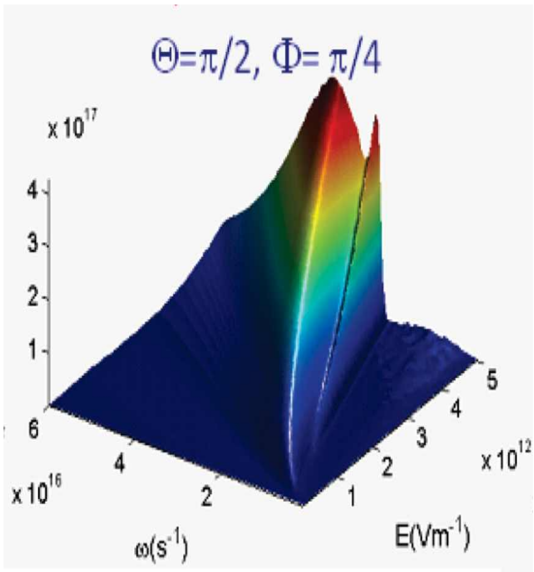
Figure 4.5: Differential ionization rate $dw/d\Omega$ for circular polarized laser field, $\alpha = \beta = 1/\sqrt{2}$ at different detection directions Θ and Φ.

In our calculation, we have generalize the Keldysh's formalism for arbitrary momentum of the photoelectron whereby Keldysh has neglected the higher order term of the momentum. Fig. 4.7 has shown the comparison between my result with Keldysh's result. As we can see, the photoionization rate is totally agree at the low frequency regime (small momentum due to the proportionality of the frequency and momentum). However, the photoionization rate is totally disagree at the high frequency regime which is higher momentum for the photoelectron.

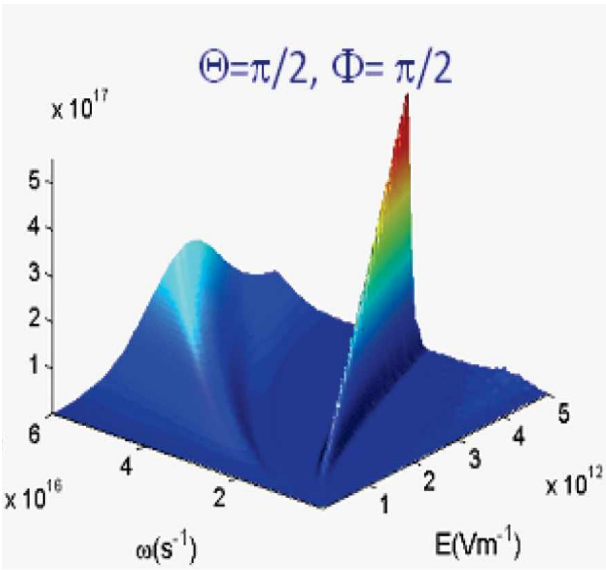
To conclude, the Volkov wavefunction and Keldysh's formalism is studied as in the previous section. We have obtained the semi-analytical expressions for photoionization



(a) Detection direction $\Theta = \pi/2$ and $\Phi = 0$.



(b) Detection direction $\Theta = \pi/2$ and $\pi/4$.



(c) Detection direction $\Theta = \pi/2$ and $\pi/2$.

Figure 4.6: Diferential Ionization rate $dw/d\Omega$ for elliptic polarized laser field, $\alpha = 1/\sqrt{5}, \beta = 2/\sqrt{5}$ at different detection directions Θ and Φ.

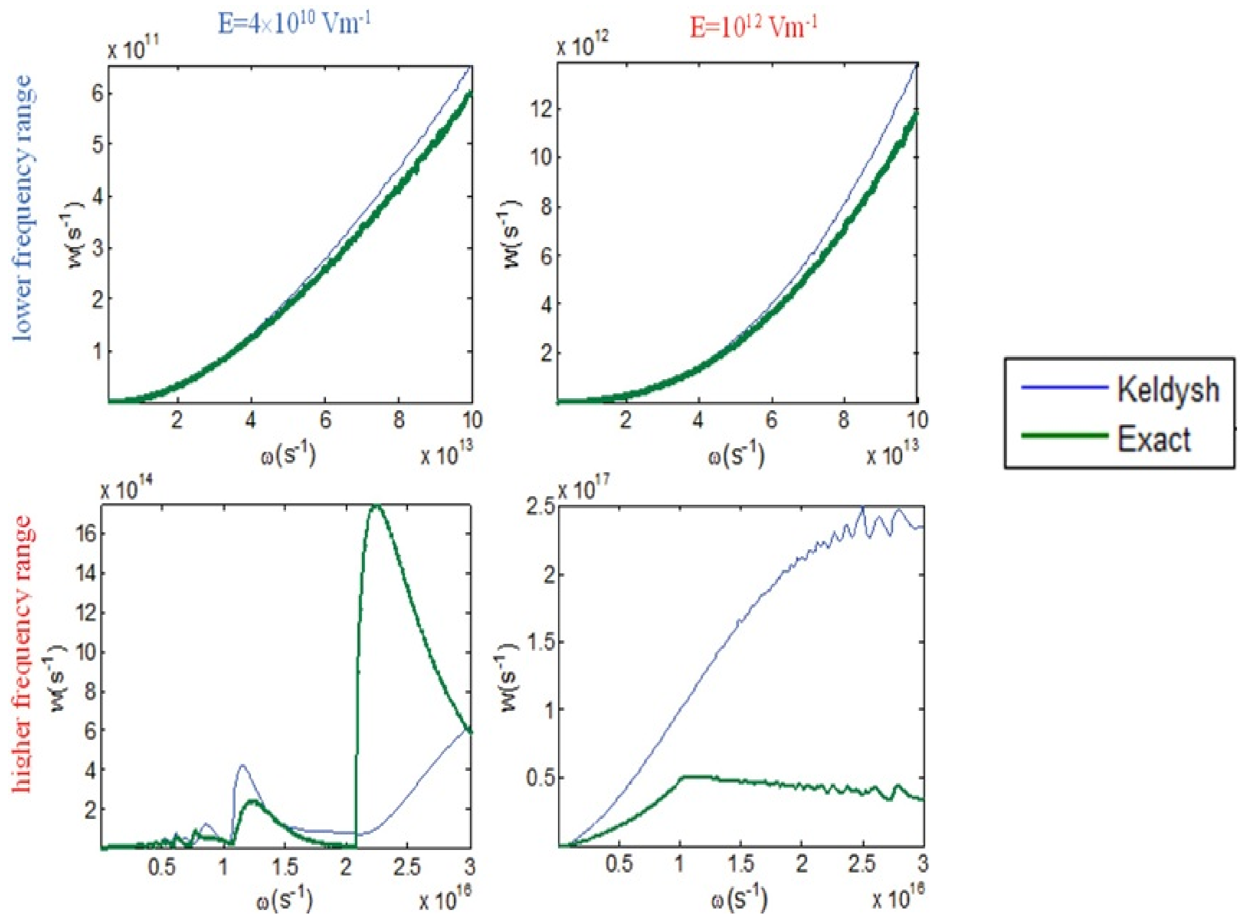


Figure 4.7: Comparison between the exact photoionization rate with Keldysh's result.

rates driven by circular and linearly polarized light. The exact result for linear polarized is qualitative different from the Keldysh's theory only at high field regime. In the low field regime, the results agree. Besides, the multiple saddle points in the linear polarized case give rise to the interference feature. Such feature also appears in the circular polarized case. We have also generalized the Keldysh's formalism for arbitrary momentum of the photoelectron. The comparison between the exact photoionization rate and Keldysh's original result has been done. The theory developed can be extended to calculate higher order terms in the perturbative formalism. For example, the second order term in the transition amplitude gives the effects of recollision and high harmonic generation (HHG) driven by circular polarized laser. The semianalytical results provide some physical insights and can be compared with the results obtained by solving the time dependent Schrodinger equation (K.-J. Yuan & Bandrauk, 2011b).

CHAPTER 5

GENERALIZED MODEL OF PHOTOIONIZATION

5.1 Introduction

Over the past few decades, a lot of significant researches and efforts have been done in order to compute a complete theoretical model to describe the photoionization rate (Zhou & Chu, 2011) of atoms by considering various aspects and factors. One of the main concern is the initial energy state problem. In previous chapter, we have further extended the Keldysh's theory by taking consideration of arbitrary momentum of the photoelectron. By using the residue theorem, the exact photoionization rate of arbitrary momenta is obtained

$$w = \frac{m}{(2\pi\hbar^2)^2} 2\pi \int_0^{\pi} \sum_{n==n_0}^{\infty} |L(\mathbf{p}_n)|^2 p_n \sin\Theta d\Theta, \tag{5.1}$$

where the ionization amplitude

$$|L(\mathbf{p}_n)|^2 = (\frac{4\hbar\omega I_0}{eE})^2 \pi a \left| \frac{e^{iS(\mathbf{p}_n,u_+)}}{\eta_+ \cos\omega t_+} + \frac{e^{iS(\mathbf{p}_n,u_-)}}{\eta_- \cos\omega t_-} \right|^2, \tag{5.2}$$

with double saddle points

$$u_{\pm} = -\gamma\chi\cos\Theta \pm \gamma\sqrt{(\chi\cos\Theta)^2 - (1+\chi^2)} \tag{5.3}$$

$$\omega t_+ = \sin^{-1} u_+, \tag{5.4}$$

$$\omega t_- = \pi - \sin^{-1} u_-, \tag{5.5}$$

and the phase for each saddle point is $\eta_{\pm} = \pm\sqrt{1+\chi^2\sin^2\Theta} + i\frac{u_{\pm}^3}{\gamma}$ and the parameter $= \frac{p_n}{\sqrt{2mI_0}} = \sqrt{\frac{\hbar\omega}{I_0}(n-n_0)}$ depends on the frequency ω and photon number n.

However, the result is restricted to the initial energy level at the ground state hydrogen atom. In this chapter, we have extented the exact model to adapt arbitrary initial energy level of hydrogen atom. A general analytical expression for arbitrary $n00$ energy level is obtained where n is the principal quantum number. Meanwhile, semianalytical expression is obtained for arbitrary nlm energy level where l is the azimuthal quantum number and m is the magnetic quantum number. We compare the features of the angular distribution for different orbital angular momentum and magnetic states. Polarization of the laser has significant effects on the ionization pattern by intense laser field.

5.2 Literature Review

In the early development of the theoretical framework, Smirnov and Chibisov (Smirnov & Chibisov, 1966) obtained an analytical result for the photoionization rate of an atom of arbitrary energy level in the year of 1965. However, they only consider the interaction of atom with laser field, but not intense laser field. Further enhancement had been done by M. Perelomov, V. S. Popov, and M. V. Terent'ev (Perelomov et al., 1966) in the later year 1966 by improving the model to take in consideration of the strong ionization of an atom in intense laser field but anyway the model is not generalized for arbitrary energy level of the atom. In the later work of M. V. Ammosov, N. B. Delone and V. P. Krainov (Ammosov, Delone, & Krainov, 1986) in 1986, their work is also known as the famous ADK theory which is the most complete analytical model of photoionization rate for arbitrary energy level of an atom in intense linearly polarized laser field. Meanwhile, in recent work of M. Protopapas, D. G. Lappas and P. L. Knight (Protopapas, Lappas, & Knight, 1997), they formulate the numerical model of strong field ionization in arbitrary laser polarizations. However, their work is restricted to numerical result only. Currently, analytical model of photoionization in strong field for arbitrary laser polarizations would a challenge for recent theoretical research.

In the following section, a new extension has been introduced into Keldysh's formalism where the theoretical model has been generalized to adopt pulsed laser as the energy source. The past models such as Keldysh, ADK and PPT's model only consider the continuous wave (CW) laser as the energy source. However, in this new era, pulsed laser such as Ti-Sa laser is a common tools for experimental usage. In spite of this, the modification on the theoretical model is a must so that a complete and general model is always ready for experimental verification. The theoretical model will be derived and discuss the result in details.

5.2.1 Pulse Envelope function

In Keldysh's work, the formalism of photoionization rate was introduced with the continuous wave (CW) laser as the energy source.

$$\mathbf{E} = \mathbf{E}_0 \cos(\omega t + \varphi) \tag{5.6}$$

However in this paper, we consider the pulse laser as the main energy source by applying modification on the external electric field with a pulse envelope function, it may define as

$$\mathbf{E}_{lipulse} = E_0 g(t) \cos(\omega t + \varphi) \widehat{\mathbf{z}} \tag{5.7}$$

for linear polarized laser field and

where $g(t)$ may define as a general function for pulse envelope, i.e. Gaussian pulse envelope, Lorentzian pulse envelope, etc.

The vector potential, $\mathbf{A}_l$ of the linear polarized laser field can be obtained via the expression $\mathbf{E} = -\frac{d\mathbf{A}}{dt}$. After perform the integration by part, we get the vector potential with the following expression

$$\mathbf{A}_l = -E_0 \mathbf{h}_l(t) \tag{5.8}$$

where $h(t)$ is general function of vector potential with respect of t

$$\mathbf{h}_l(t) = \left[\frac{g(t)}{\omega} \sin(\omega t + \varphi) - \int \frac{dg(t)}{dt} \frac{\sin(\omega t + \varphi)}{\omega} dt \right] \widehat{\mathbf{z}} \tag{5.9}$$

We notice that when the pulse envelope function is a constant, say $g(t) = 1$ and the phase difference $\varphi = 0$, then we have $\frac{dg(t)}{dt} = 0$ and the vector potential is reduced to the continuous wave laser case as in our previous work (R. Ooi et al., 2012).

$$\mathbf{A} = -\frac{\mathbf{E}_0}{\omega} \sin(\omega t + \varphi) \widehat{\mathbf{z}} \tag{5.10}$$

Meanwhile, for circular polarized laser field, the electric field will be redefined as

$$\mathbf{E}_{cirpulse} = E_0 g(t) \left[\alpha \cos(\omega t + \varphi) \widehat{\mathbf{x}} + \beta \sin(\omega t + \vartheta) \widehat{\mathbf{y}} \right] \tag{5.11}$$

By using the same equation $\mathbf{E} = -\frac{d\mathbf{A}}{dt}$, we can obtain the vector potential of the circular polarized electric field,

$$\mathbf{A}_{cir} = -E_0 \mathbf{h}_{cir}(t) \tag{5.12}$$

where

$$h(t) = \frac{g(t)}{\omega} \sin(\omega t + \varphi) - \int \frac{dg(t)}{dt} \frac{\sin(\omega t + \varphi)}{\omega} dt \tag{5.13}$$

5.2.2 New Volkov state

Since the new vector potential includes the pulse envelope, hence the minimal coupling of the Schrödinger equation is also being modified,

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \Psi(\mathbf{r},t) &= \frac{1}{2m} \left(\frac{\hbar}{i} \nabla - e\mathbf{A} \right)^2 \Psi(\mathbf{r},t) \\ &= \frac{1}{2m} \left(-\hbar^2 \nabla^2 + e^2 \mathbf{A}^2 - \frac{\hbar}{i} \nabla \cdot \mathbf{A} - \frac{\hbar}{i} e\mathbf{A} \cdot \nabla \right) \Psi(\mathbf{r},t) \end{aligned} \tag{5.14}$$

Consequently, the Volkov wavefunction is obtained

$$\Psi(\mathbf{r},t) = \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar} \right) \exp\left[-\frac{i}{\hbar} \int \frac{1}{2m} (\mathbf{p} - e\mathbf{A})^2 dt \right] \tag{5.15}$$

By applying the length gauge transformation, the Volkov wavefunction can be obtained

$$\begin{aligned} \Psi_p(\mathbf{r},t) &= \exp\left\{ \frac{i}{\hbar} \left[\Pi(t) \cdot \mathbf{r} - \int \frac{1}{2m} \Pi(\tau)^2 d\tau \right] \right\} \\ &= f(\mathbf{r},t)\, b(t) \end{aligned} \tag{5.16}$$

where $\Pi(t)$ is defined as

$$\Pi(t) = \mathbf{p} - e\mathbf{A}(t) \tag{5.17}$$

and the vector potential

$$\mathbf{A} = -\mathbf{E}_0 \left[\frac{g(t)}{\omega} \sin(\omega t + \varphi) - \int \frac{dg(t)}{dt} \frac{\sin(\omega t + \varphi)}{\omega} dt \right] \tag{5.18}$$

5.3 Linear Polarized Pulsed Laser

In the first case, let us analyze the ionization rate of the linear polarized pulse laser field via

$$\mathbf{E}_{lipulse} = E_0 g(t) \cos(\omega t + \varphi) \widehat{\mathbf{z}} \tag{5.19}$$

5.3.1 Matrix Element Prefactor

The transition of the electron of hydrogen atom from arbitrary energy level, ψ_{nlm} to Volkov state, $\Psi_p(\mathbf{r},t)$ under the interaction of pulse laser source, E_{pulse} can be described by computing the matrix element prefactor. In the linear polarized laser field, the general matrix element prefactor is written as

$$\begin{aligned} V_{0p}(\mathbf{p},t) &= \int \Psi^*_{\mathrm{P}rel}(\mathbf{r},t) e\mathbf{E}_{lipulse} \cdot \mathbf{r} \Psi_{nlm} d^3 r & (5.20) \\ &= \psi_{nlm}(t) b^*(t) g(t) \cos(\omega t + \varphi) \int \psi_{nlm}(\mathbf{r}) f^*(\mathbf{r},t) e E_0 r \cos\theta d^3 r \\ &= \psi_{nlm}(t) b^*(t) g(t) \cos(\omega t + \varphi) V_0(\Pi(t)) & (5.21) \end{aligned}$$

In order to simplify the initial calculation, we set the special case for hydrogen ground state

$$\begin{aligned} V_{0p}(\mathbf{p},t) &= \int \Psi^*_{\mathrm{P}rel}(\mathbf{r},t) e\mathbf{E}_{lipulse} \cdot \mathbf{r} \Psi_{nlm}(\mathbf{r},t) d^3 r & (5.22) \\ &= \psi_s(t) b^*(t) g(t) \cos(\omega t + \varphi) \int \psi_{nlm}(\mathbf{r}) f^*(\mathbf{r},t) e E_0 r \cos\theta d^3 r \\ &= \psi_s(t) b^*(t) g(t) \cos(\omega t + \varphi) V_0(\Pi(t)) & (5.23) \end{aligned}$$

$$\Psi_{nlm}(\mathbf{r},t) = \psi_{nlm}(\mathbf{r}) \psi_{nlm}(t) \tag{5.24}$$

Since pulse has a large bandwidth we might have to include the first few excited states in addition to the ground state. So the initial wavefunction should be a superposition of a few states,

$$\Psi_{nlm}(\mathbf{r},t) =_g C_g \psi_g(\mathbf{r},t) \tag{5.25}$$

where $V_0(\Pi(t))$ is the general transition matrix

$$V_0(\Pi(t)) = \int \psi_{nlm}(\mathbf{r}) \exp\left\{-\frac{i}{\hbar}\Pi(t)\cdot\mathbf{r}\right\} eE_0 r\cos\theta d^3r \tag{5.26}$$

$$= A\int r^l e^{-r/na_0}\left[L_{n-l-1}^{2l+1}\left(\frac{2r}{na_0}\right)\right] e^{im\phi} P_l^m(\cos\theta) \exp\left\{-\frac{i}{\hbar}\Pi(t)\cdot\mathbf{r}\right\} r\cos\theta d^3r$$

where $A = eE_0\left(\frac{2}{na_0}\right)^l \sqrt{\left(\frac{2}{na_0}\right)^3 \frac{(n-l-1)!}{2n[(n+1)!]^3}}\sigma\sqrt{\frac{(2l+1)}{4\pi}\frac{(l-|m|)!}{(l+|m|)!}}$ and $\Pi(t) = \mathbf{p} - e\mathbf{A}(t)$.

In order to simplify the matrix element prefactor, we make an assumption that the direction of polarization is just parallel to the electric field $\mathbf{E}$

$$\mathbf{A}(t) = -E_0\left[\frac{g(t)}{\omega}\sin(\omega t+\varphi) - \int \frac{dg(t)}{dt}\frac{\sin(\omega t+\varphi)}{\omega}dt\right]\widehat{\mathbf{z}} \tag{5.27}$$

and the dot product yields

$$\mathbf{A}(t)\cdot\mathbf{r} = -E_0\left[\frac{g(t)}{\omega}\sin(\omega t+\varphi) - \int \frac{dg(t)}{dt}\frac{\sin(\omega t+\varphi)}{\omega}dt\right]\cdot r(\cos\theta)(\widehat{\mathbf{z}}\cdot\widehat{\mathbf{z}})$$

$$= -E_0\left[\frac{g(t)}{\omega}\sin(\omega t+\varphi) - \int \frac{dg(t)}{dt}\frac{\sin(\omega t+\varphi)}{\omega}dt\right] r\cos\theta \tag{5.28}$$

Similarly, we use the classical approximation that assume only z-momentum part p_z contribute since $\mathbf{p}$ is parallel to $\mathbf{E}$

$$\Pi(t)\cdot\mathbf{r} = [\mathbf{p} - e\mathbf{A}(t)]\cdot\mathbf{r} \tag{5.29}$$

$$= \left\{E_0\left[\frac{g(t)}{\omega}\sin(\omega t+\varphi) - \int \frac{dg(t)}{dt}\frac{\sin(\omega t+\varphi)}{\omega}dt\right] + p_z\right\} r\cos\theta \tag{5.30}$$

Hence, we reduce the matrix element prefactor

$$V_0(\Pi(t)) = \int \psi_{nlm}(\mathbf{r}) \exp\left\{-\frac{i}{\hbar}\Pi(t)\cdot\mathbf{r}\right\} eE_0 r\cos\theta d^3r \tag{5.31}$$

$$= A\int_0^\infty\int_0^\pi\int_0^{2\pi} r^l e^{-r/na_0}\left[L_{n-l-1}^{2l+1}\left(\frac{2r}{na_0}\right)\right] e^{im\phi} P_l^m(\cos\theta) \tag{5.32}$$

$$\times \exp\left\{-\frac{i}{\hbar}\Pi(t)\cdot\mathbf{r}\right\} r\cos\theta r^2\sin\theta d\phi d\theta dr$$

$$= A\int_0^{2\pi} F(\theta,t) e^{im\phi} d\phi \tag{5.33}$$

where

$$F(\theta,t)=\int_0^{\infty}\int_0^{\pi} r^{l+3}e^{-r/na_0}\exp\left\{-\frac{i}{\hbar}\Pi(t)\cdot\mathbf{r}\right\}\left[L_{n-l-1}^{2l+1}\left(\frac{2r}{na_0}\right)\right]P_l^m(\cos\theta)\cos\theta\sin\theta d\theta dr \tag{5.34}$$

By refering to the Eq. 5.33, we note that for any other value of $m\neq 0$,

$$\begin{aligned}\int_0^{2\pi}F(r,\theta)e^{im\phi}d\phi &= F(\theta,t)\left|\frac{e^{im\phi}}{im}\right|_0^{2\pi} \\ &= F(\theta,t)\left|\frac{\cos m\phi+i\sin m\phi}{im}\right|_0^{2\pi} \\ &= 0\end{aligned} \tag{5.35}$$

However, for the case $m=0$, we obtain the following expression

$$\int_0^{2\pi}F(\theta,t)e^{im\phi}d\phi=2\pi F(\theta,t) \tag{5.36}$$

5.3.1 (a) General solution for energy level n00

Equation 5.33 will give the value of 2π only for $m=0$. Hence, from here we can conclude that in linear polarized pulsed laser field, m can only take value of 0 since there is no any circular or elliptical moment of the electron. The matrix element becomes

$$V_0(\Pi(t)) = 2\pi F(\theta,t) \tag{5.37}$$

$$= 2\pi A\int_0^{\infty}\int_0^{\pi} r^{l+3}e^{-r/na_0}\exp\left\{-\frac{i}{\hbar}\Pi(t)\cdot\mathbf{r}\right\}\left[L_{n-l-1}^{2l+1}\left(\frac{2r}{na_0}\right)\right] \times P_l^m(\cos\theta)\cos\theta\sin\theta d\theta dr \tag{5.38}$$

The semi-analytical function $F(\theta,t)$ can be solved exactly if we set a condition so that l can take the value of 0 only. Consequently,

$$F(\theta,t)=\int_0^{\infty}\int_0^{\pi} r^3e^{-r/na_0}\exp\{-iBr\}\left[L_{n-1}^{1}\left(\frac{2r}{na_0}\right)\right]\cos\theta\sin\theta d\theta dr \tag{5.39}$$

with $B=\frac{1}{\hbar}\left\{E_0\left[\frac{g(t)}{\omega}\sin(\omega t+\varphi)-\int\frac{dg(t)}{dt}\frac{\sin(\omega t+\varphi)}{\omega}dt\right]+p_z\right\}\cos\theta$.

In order to get an analytical solution for $F(\theta,t)$, we perform a little trick to make a transformation on r, say $x = \frac{2r}{na_0}$, hence

$$r = \frac{na_0 x}{2}, \tag{5.40}$$

$$b = \frac{na_0 B}{2}, \tag{5.41}$$

then we have

$$\begin{aligned}
F(\theta,t) &= \int_0^\infty \int_0^\pi r^3 e^{-r/na_0} \exp\{-iBr\} \left[L_{n-1}^1\left(\frac{2r}{na_0}\right)\right] \cos\theta \sin\theta d\theta dr \qquad (5.42)\\
&= \left(\frac{na_0}{2}\right)^4 \int_0^\infty \int_0^\pi e^{-x/2} \left[L_{n-1}^1(x)\right] x^3 \exp[-ibx] \cos\theta \sin\theta d\theta dx\\
&= -\left(\frac{na_0}{2}\right)^4 \int_0^\pi \frac{32n\left(-1+3n^2a_0^2B^2-2n^2+6in^2a_0B\right)(-1+ina_0B)^{n-3}}{(1+ina_0B)^{n+3}} \qquad (5.43)\\
&\quad \times \cos\theta \sin\theta d\theta.
\end{aligned}$$

Hence, the matrix element is simplified as

$$V_0(\Pi(t)) = -2\pi \left(\frac{na_0}{2}\right)^4 A \int_0^\pi \frac{32n\left(-1+3n^2a_0^2B^2-2n^2+6in^2a_0B\right)(-1+ina_0B)^{n-3}}{(1+ina_0B)^{n+3}} \cos\theta \sin\theta d\theta. \tag{5.44}$$

Eq. 5.44 is quite complicated with the θ term, let make another simplification on B

$$\begin{aligned}
B &= \frac{1}{\hbar}\left\{E_0\left[\frac{g(t)}{\omega}\sin(\omega t+\varphi) - \int \frac{dg(t)}{dt}\frac{\sin(\omega t+\varphi)}{\omega}dt\right] + p_z\right\}\cos\theta\\
&= D\cos\theta,
\end{aligned}$$

with $D = \frac{1}{\hbar}\left\{E_0\left[\frac{g(t)}{\omega}\sin(\omega t+\varphi) - \int \frac{dg(t)}{dt}\frac{\sin(\omega t+\varphi)}{\omega}dt\right] + p_z\right\}$.

Let us introduce a dimensionless quantity,

$$X = \cos\theta, \tag{5.45}$$

and the differentiation of it yields

$$\frac{dX}{d\theta} = -\sin\theta.$$

Hence, by making a substitution in to the equation 5.44, we obtain the analytical solution of the matrix element prefactor for the electron energy level $n00$.

$$\begin{aligned} V_0(\Pi(t)) &= -2\pi\left(\frac{na_0}{2}\right)^4 A\int_0^\pi \frac{32n\left(-1+3n^2a_0^2B^2-2n^2+6in^2a_0B\right)\left(-1+ina_0B\right)^{n-3}}{\left(1+ina_0B\right)^{n+3}} \qquad (5.46)\\ &\quad \times\cos\theta\sin\theta d\theta \\ &= 2\pi\left(\frac{na_0}{2}\right)^4 A\int_0^\pi \frac{32n\left(-1+3n^2a^2D^2X^2-2n^2+6in^2aDX\right)\left(-1+inaDX\right)^{n-3}}{\left(1+inaDX\right)^{n+3}}XdX \\ &= 64n\pi\left(\frac{na_0}{2}\right)^4 A\left[-\frac{(-1)^n}{2a^2D^2n^2}+\frac{\left(\frac{i+aDn\pi}{-i+aDn\pi}\right)^n\left(1+2iaDn^2\pi+3a^2D^2n^2\pi^2\right)}{2\left(naD+a^3D^3n^3\pi^2\right)^2}\right]. \qquad (5.47) \end{aligned}$$

5.4 $L(\mathbf{p},t)$ function and the action part

Next, we redefine the entire function of matrix element prefactor $V_{0p}(\mathbf{p},t)=\int\Psi^*_{\mathrm{P}rel}(\mathbf{r},t)\,e\mathbf{E}\cdot\mathbf{r}\Psi_s(\mathbf{r},t)\,d^3r$ as $L(\mathbf{p},t)\cos(\omega t+\varphi)$, hence

$$\begin{aligned} L(\mathbf{p},t) &= V_0(\Pi(t))\,\psi_s(t)\,b^*(t)\,g(t) \qquad (5.48)\\ &= V_0(\Pi(t))\,g(t)\,e^{iS(\mathbf{p},t)}, \end{aligned}$$

where the $S(\mathbf{p},t)$ function is the action of the transition process

$$S(p,t)=-\frac{1}{\hbar}\int_0^t\left[I_n-e\mathbf{E}_{lipulse}\rho-\frac{1}{2}e^2\mathbf{E}^2_{lipulse}-\sqrt{\left(\mathbf{p}-e\mathbf{A}_l\right)^2c^2+m^2c^4}+mc^2\right]dt, \qquad (5.49)$$

with

$$\begin{aligned} I_n &= \frac{I_0}{n^2}, \qquad (5.50)\\ \mathbf{E}_{lipulse} &= E_0 g(t)\cos(\omega t+\varphi)\,\widehat{\mathbf{z}}, \qquad (5.51)\\ \mathbf{A}_l &= -E_0\mathbf{h}_l(t), \qquad (5.52)\\ \mathbf{h}_l(t) &= \left[\frac{g(t)}{\omega}\sin(\omega t+\varphi)-\int\frac{dg(t)}{dt}\frac{\sin(\omega t+\varphi)}{\omega}dt\right]\widehat{\mathbf{z}}, \qquad (5.53) \end{aligned}$$

where $g(t)$ is arbitrary pulse envelope function such as lorentzian function, gaussian function, etc.

Next, we introduce another function to replace the matrix element prefactor,

$$L(\mathbf{p}) = V_{0p}(\mathbf{p},t) \tag{5.54}$$

$$= V_0(\Pi(t))\, g(t)\cos(\omega t+\varphi) \tag{5.55}$$

$$\times \exp\left\{\frac{i}{\hbar}\left[\Omega(\mathbf{p}) - \frac{e^2E_0^2}{4m\omega^2}t + \frac{e(\mathbf{p}\cdot\mathbf{E}_0)}{m}\left[\int_0^t h(t)\,dt\right] + \frac{e^2E_0^2}{2m}\left[\int_0^t h(t)^2\,dt\right]\right]\right\}.$$

By applying fourier transformation on $L(\mathbf{p})$ function

$$L(p_n) = \frac{1}{2\pi}\int_{-T/2}^{T/2} L(\mathbf{p})\exp\left\{\frac{i}{\hbar}\left[n\hbar\omega - \Omega(\mathbf{p})\right]t\right\}dt \tag{5.56}$$

$$= \frac{1}{2\pi}V_0(\Pi(t))\, g(t)\cos(\omega t+\varphi) \tag{5.57}$$

$$\times \exp\left\{\frac{i}{\hbar}\left[n\hbar\omega t - \frac{e^2E_0^2}{4m\omega^2}t + \frac{e(\mathbf{p}_n\cdot\mathbf{E}_0)}{m}\left[\int_0^t h(t)\,dt\right] + \frac{e^2E_0^2}{2m}\left[\int_0^t h(t)^2\,dt\right]\right]\right\}dt.$$

The conservation of energy is satisfied by the expression $n\hbar\omega = \Omega(\mathbf{p}) = I_0 + K + U_p$, where I_0 is the ionization potential, $K = \frac{p^2}{2m}$ is the kinetic energy of the photoelectron and $U_p = \frac{e^2E_0^2}{4m\omega^2}$ is the ponderomotive energy.

The contour integration $V_0(\Pi(t))\, g(t)\cos(\omega t+\varphi)\, e^{iS(\mathbf{p}_n,t)}dt$ will be solved fully numerically, and finally the semi-analytical expression of the photoionization rate of pulse laser yields

$$w = \frac{m}{2\pi\hbar^4}\int_0^{\pi}\sum_{n=n_0}^{\infty}\left|L(p_n)\right|^2 p_n \sin\Theta d\Theta. \tag{5.58}$$

5.4.1 General Rate Of Elliptical Polarized Field

For a hydrogenic atom in an elliptical polarized intense laser field $\mathbf{E} = E(\alpha\cos\omega t, \beta\sin\omega t, 0)$ where the coefficient α and β determine the ellipticity $\varepsilon = \alpha/\beta$ of the laser field, the general photoionization rate of (as shown in our previous result) is defined as:

$$w = \frac{m}{\left(2\pi\hbar^2\right)^2}\int_0^{2\pi}\int_0^{\pi}\sum_{n=n_0'}^{\infty}\left|L(\mathbf{p}_n)\right|^2 p_n \sin\Theta d\Theta d\Phi, \tag{5.59}$$

where

$$L(\mathbf{p}_n) = \frac{1}{2\pi}\frac{V_0(\Pi_n(u))}{\sqrt{1-u^2}}e^{iS(\mathbf{p}_n,u)}du \tag{5.60}$$

$$= \frac{1}{2\pi}\int_{-\pi}^{\pi} V_0(\Pi(s))\exp iS(\mathbf{p}_n,s)\,ds,$$

with the transition matrix element $V_0(\Pi(s))$ represents the transition of the photoelectron from initial state $\psi_s(\mathbf{r})$ to the continuum Volkov state $\psi_p(\mathbf{r},t) = \exp\left\{\frac{i}{\hbar}\left[\Pi(t)\cdot\mathbf{r} - \int_0^t \frac{\Pi(\tau)^2}{2m} d\tau\right]\right\}$ with $\Pi(\tau) = \mathbf{p} - e\mathbf{A}(t)$,

$$V_0(t) = e\int \psi_s(\mathbf{r}) Er\sin\theta C(t,\phi) e^{-i\Xi} r^2 dr \sin\theta d\theta d\phi \tag{5.61}$$

Meanwhile, $S(\mathbf{p}_n, s)$ represents the action phase during the photoionization,

$$S(\mathbf{p},s) = ns - \frac{U_p}{\hbar\omega}\frac{\left(\alpha^2-\beta^2\right)\sin 2s}{2} - \frac{eE}{\hbar m\omega^2}\left[\alpha p_x \cos s + \beta p_y \sin s\right] \tag{5.62}$$

with $u = \sin s$ and $s = \omega t$.

However, the angular dependence of the photoionization rate is obtained by differentiate Eq. 5.59 with respect of the polar angle Θ and azimuthal angle Φ

$$\frac{dw}{d\Omega_a} = \frac{m}{\left(2\pi\hbar^2\right)^2} \sum_{n=n_0}^{\infty} \left|L(\mathbf{p}_n)\right|^2 p_n. \tag{5.63}$$

5.4.1 (a) Volkov State in Elliptical Polarized Laser Field

For a hydrogenlike atom which is placed in the elliptical polarized laser field as defined as following

$$\mathbf{E} = E(\alpha\cos\omega t, \beta\sin\omega t, 0). \tag{5.64}$$

In this case, the elliptical polarized laser is located in *xy*-plane for simplicity purpose where the coefficient α and β determine the ellitipcity of the laser field, where

$$\varepsilon = \alpha/\beta. \tag{5.65}$$

When the ellipticity ratio $\varepsilon = 1$, the laser field will result a circularly polarized laser field which satisfy the condition $\alpha = \beta = 1$. To obtain a vector potential $A(t)$ for the elliptical polarized laser field above, firstly we have to make sure that the Maxwell's equation is satisfied

$$\mathbf{E} = -\frac{\partial\mathbf{A}(t)}{\partial t} - \nabla\phi, \tag{5.66}$$

$$\mathbf{B} = \nabla \times \mathbf{A}(t). \tag{5.67}$$

The dot product of the electric field is defined as

$$\nabla \cdot \mathbf{E} = -\frac{\partial}{\partial t}\nabla \cdot \mathbf{A}(t) - \nabla^2 \phi. \tag{5.68}$$

Since this process is an electromagnetic field interaction, hence we also have to consider the Coulomb gauge for this interaction, where $\nabla \cdot \mathbf{A} = 0$ and hence imply that the vector potential expression to be

$$\begin{aligned} \mathbf{A}(t) &= -\int_{-\infty}^{t} \mathbf{E}(t')\, dt' \\ &= -\int_{-\infty}^{t} E(\alpha \cos \omega t \widehat{\mathbf{x}} + \beta \sin \omega t \widehat{\mathbf{y}})\, dt' \\ &= \frac{E}{\omega}(-\alpha \sin \omega t \widehat{\mathbf{x}} + \beta \cos \omega t \widehat{\mathbf{y}}). \end{aligned} \tag{5.69}$$

For such an intense laser field, the electron of the particular hydrogenic atom will be excited to the continuum level. Hence, by applying the minimal coupling, the system is described by the Schrödinger's equation as below

$$\begin{aligned} i\hbar \frac{\partial}{\partial t}\Psi(r,t) &= \frac{1}{2m}\left(\frac{\hbar}{i}\nabla - e\mathbf{A}(t)\right)^2 \Psi(r,t) \\ &= \frac{1}{2m}\left(\frac{\hbar}{i}\nabla + \frac{eE}{\omega}(\alpha \sin \omega t \widehat{\mathbf{x}} - \beta \cos \omega t \widehat{\mathbf{y}})\right)^2 \Psi(r,t). \end{aligned} \tag{5.70}$$

Next, the equation 5.70 can be simplified by using the separation of variables method, this imply that

$$\Psi(r,t) = \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)(t), \tag{5.71}$$

where $\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)$ is the spatial dependence part meanwhile (t) is a function of time dependent. We substitute the equation 5.84 into equation 5.70 and yield

$$i\hbar \frac{\partial}{\partial t} \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)(t) = \frac{1}{2m}\left[\frac{\hbar}{i}\nabla - e\mathbf{A}(t)\right]^2 \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)(t) \tag{5.72}$$

$$i\hbar \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\frac{\partial}{\partial t}(t) = \frac{1}{2m}\left[-\hbar^2\nabla^2 - e\mathbf{A}(t)\right]^2 \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)(t) \tag{5.73}$$

5.4.1 (b)   Initial State with Arbitrary n-Energy Level

In this chapter, the general hydrogen wavefunction is considered, in which is defined as following

$$\Psi_{n,l,m}(\mathbf{r}) = \sqrt{\left(\frac{2}{na_0}\right)^3 \frac{(n-l-1)!}{2n[(n+1)!]^3}} R_{nl}(\mathbf{r}) Y_l^m(\theta,\phi) \tag{5.74}$$

with the radial wavefunction is defined as

$$R_{nl} = e^{-r/na} \left(\frac{2r}{na_0}\right)^l \left[L_{n-l-1}^{2l+1}\left(\frac{2r}{na_0}\right)\right] \tag{5.75}$$

and the angular wavefunction is defined as

$$Y_l^m(\theta,\phi) = \sigma\sqrt{\frac{(2l+1)}{4\pi}\frac{(l-|m|)!}{(l+|m|)!}} e^{im\phi} P_l^m(\cos\theta) \tag{5.76}$$

where $L_n^{(a)}(x) = {}_{j=0}^{\ n}(-1)^j \begin{pmatrix} n+a \\ n-j \end{pmatrix} \frac{x^j}{j!}$ is the associated Laguarre polynomials,

$\sigma = \left\{ \begin{array}{c} \sigma = (-1)^m \text{ if } m \geq 0 \\ \sigma = 1 \text{ if } m < 0 \end{array} \right\}$ is the piecewise function,

$P_l^m(x) = \frac{(-1)^m}{2^l l!}\left(1-x^2\right)^{m/2} \frac{d^{l+m}}{dx^{l+m}}\left(x^2-1\right)^l$ is the associated Legendre polynomials.

Hence, the transition matrix element of the initial state of arbitrary energy level to the continuum Volkov state is redefined as

$$\begin{aligned} V_0(\Pi(t)) &= \int_0^\infty \int_0^\pi \int_0^{2\pi} \exp\left(-\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{r}\right) \exp\left(-\frac{i}{\hbar}\frac{e\mathbf{E}\cdot\mathbf{r}}{\omega}\sin\omega t\right) e\mathbf{E}\cdot\mathbf{r}\Psi_{n,l,m}(\mathbf{r}) r^2 \sin\theta d\phi d\theta dr \\ &= A\int_0^\pi \int_0^{2\pi} W(r) C(t,\phi) \sin^2\theta d\phi d\theta, \end{aligned} \tag{5.77}$$

where $A = eE\left(\frac{2}{na_0}\right)^l \sqrt{\left(\frac{2}{na_0}\right)^3 \frac{(n-l-1)!}{2n[(n+1)!]^3}} \sigma \sqrt{\frac{(2l+1)}{4\pi}\frac{(l-|m|)!}{(l+|m|)!}}$ is the coefficient factor varying with the different n-energy level, $C(t,\phi) = (\alpha\cos\omega t\cos\phi + \beta\sin\omega t\sin\phi) e^{im\phi}$ is the function of the laser pulse time, t and the azimuthal angle ϕ, and

$W(r) = \int_0^\infty \left\{e^{-r/na_0}\left[L_{n-l-1}^{2l+1}\left(\frac{2r}{na_0}\right)\right]\right\} r^{l+3} \exp[-iBr]\, dr$ is the integration over the radial part that consists of

$$B = \frac{1}{\hbar}\left(Q\sin\theta + P\cos\theta\right) \quad (5.78)$$

$$Q = \left(p_x + \alpha\frac{eE}{\omega}\sin\omega t\right)\cos\phi + \left(p_y - \beta\frac{eE}{\omega}\cos\omega t\right)\sin\phi \quad (5.79)$$

$$P = p_z \quad (5.80)$$

5.5 Generalized formalism

In this section, we will formulate the model as general as possible by considering the arbitrary initial state of hydrogen atom. The initial state is playing the role of the starting point where it interacts with the laser source at the first place, thus it depends on the quantum number *nlm*. We start with the Schrödinger equation as following

$$i\hbar\frac{\partial}{\partial t}\Psi_{nlm}(\mathbf{r},t) = \left[-\frac{1}{2m}\hbar^2\nabla^2 + V(\mathbf{r}) - e\mathbf{E}\rho - \frac{1}{2}e^2\mathbf{E}^2\right]\Psi_{nlm}(\mathbf{r},t) \quad (5.81)$$

where the coefficients ρ and describe the Stark shift effect in the external electric field as defined in the work of (Kim & Cho, 2000)

$$\rho = \sum_{J=1/2}^{3/2}\kappa_0(J)\sum_k\left\langle r\left(6S_{1/2},kP_J\right)\right\rangle^2\left(\frac{1}{\omega_{6S_{1/2}} - \omega_{kP_J} + \omega_1} + \frac{1}{\omega_{6S_{1/2}} - \omega_{kP_J} - \omega_2}\right), \quad (5.82)$$

with $\kappa_0\left(\frac{1}{2}\right) = \frac{1}{9}$ and $\kappa_0\left(\frac{3}{2}\right) = \frac{2}{9}$, and

$$= \sum_{J=1/2}^{3/2}\kappa_1(J)\sum_k\left\langle r\left(6S_{1/2},kP_J\right)\right\rangle^2\left(\frac{1}{\omega_{6S_{1/2}} - \omega_{kP_J} + \omega_1} + \frac{1}{\omega_{6S_{1/2}} - \omega_{kP_J} - \omega_2}\right), \quad (5.83)$$

with $\kappa_1\left(\frac{1}{2}\right) = \frac{1}{9}$ and $\kappa_1\left(\frac{3}{2}\right) = -\frac{1}{9}$.

Next, by using the separation of variables, let

$$\Psi_{nlm}(\mathbf{r},t) = \psi_{nlm}(\mathbf{r})\,\psi_{nlm}(t) \quad (5.84)$$

We insert equation 5.84 into equation B.1, the Schrödinger equation becomes,

$$i\hbar\frac{\partial}{\partial t}\psi_{nlm}(\mathbf{r})\,\psi_{nlm}(t) = \left[-\frac{1}{2m}\hbar^2\nabla^2 + V(\mathbf{r}) - e\mathbf{E}\rho - \frac{1}{2}e^2\mathbf{E}^2\right]\psi_{nlm}(\mathbf{r})\,\psi_{nlm}(t) \quad (5.85)$$

In hyperfine model, it is well known that

$$\left[-\frac{1}{2m}\hbar^2\nabla^2+V(\mathbf{r})\right]\psi_{nlm}(\mathbf{r})=I_n\psi_{nlm}(\mathbf{r}) \tag{5.86}$$

$$\begin{aligned} I_n &= \left[\frac{m_e}{2\hbar^2}\left(\frac{e^2}{4\pi\varepsilon_0}\right)^2\right]\frac{1}{n^2} \\ &= \frac{I_0}{n^2} \end{aligned} \tag{5.87}$$

I_n is the ionization energy that varies with the energy level, n. For instance, at the hydrogen level ground state where $n=0$, the ionization energy is $I_0=13.6eV$. Hence, equation 5.85 becomes,

$$i\hbar\frac{\partial}{\partial t}\psi_{nlm}(t)=\left[I_n-e\mathbf{E}\rho-\frac{1}{2}e^2\mathbf{E}^2\right]\psi_{nlm}(t), \tag{5.88}$$

and

$$\frac{1}{\psi_{nlm}(t)}\frac{\partial}{\partial t}\psi_{nlm}(t)=-\frac{i}{\hbar}\left(I_n-e\mathbf{E}\rho-\frac{1}{2}e^2\mathbf{E}^2\right).$$

Next, by solving the differential equation,

$$\int\frac{d\psi_{nlm}(t)}{\psi_{nlm}(t)}=-\frac{i}{\hbar}\int\left(I_n-e\mathbf{E}\rho-\frac{1}{2}e^2\mathbf{E}^2\right)dt, \tag{5.89}$$

following by

$$\ln\psi_{nlm}(t)=-\frac{i}{\hbar}\left[I_nt-\int\left(e\mathbf{E}\rho+\frac{1}{2}e^2\mathbf{E}^2\right)\right]dt. \tag{5.90}$$

Hence, we obtain the solution of the time dependent wavefunction

$$\psi_{nlm}(t)=\exp\left\{-\frac{i}{\hbar}\left[I_nt-\int\left(e\mathbf{E}\rho+\frac{1}{2}e^2\mathbf{E}^2\right)\right]dt\right\}. \tag{5.91}$$

Meanwhile, the spatial part of the hydrogen general wavefunction may define as,

$$\psi_{nlm}(\mathbf{r})=\sqrt{\left(\frac{2}{na_0}\right)^3\frac{(n-l-1)!}{2n\left[(n+1)!\right]^3}}R_{nl}(\mathbf{r})Y_l^m(\theta,\phi). \tag{5.92}$$

The radial wavefunction 5.92 is consisted of radial wavefunction which is defined as

$$R_{nl} = e^{-r/na_0} \left(\frac{2r}{na_0} \right)^l \left[L_{n-l-1}^{2l+1} \left(\frac{2r}{na_0} \right) \right], \tag{5.93}$$

and the angular wavefunction where

$$Y_l^m (\theta, \phi) = \sigma \sqrt{\frac{(2l+1)}{4\pi} \frac{(l-|m|)!}{(l+|m|)!}} e^{im\phi} P_l^m (\cos\theta), \tag{5.94}$$

with

$L_n^{(a)}(x) = \mathop{}_{j=0}^{n} (-1)^j \begin{pmatrix} n+a \\ n-j \end{pmatrix} \frac{x^j}{j!}$ is the associated Laguarre polynomials,

$\sigma = \left\{ \begin{array}{c} \sigma = (-1)^m \text{ if } m \geq 0 \\ \sigma = 1 \text{ if } m < 0 \end{array} \right\}$ is the piecewise function,

and $P_l^m(x) = \frac{(-1)^m}{2^l l!} \left(1-x^2\right)^{m/2} \frac{d^{l+m}}{dx^{l+m}} \left(x^2-1\right)^l$ is the associated Legendre polynomials.

The indices of the wavefunction consists the principle quantum number, n, the azimuthal quantum number, l and also the magnetic quantum number, m. The modification on the intense laser atom interaction will be elaborated in the following section where we generalize our model into the relativistic case.

5.5.1 Relativistic Volkov State

The investigation of the electron of hydrogenlike atom in a more intense laser field would be interesting. Hence, for the electric field strength exceeds the limit of $\approx 10^{14} V m^{-1}$, the electron is beyond the classical limit and relativistic correction need to be done on the Schrödinger equation. In our general model, the Stark effect is not taken into consideration due to the complexity of the hyperfine splitting. However, we will extend the study of Stark effect on the photoionization in the future project.

Firstly, we define the external electric field for our case, which is a general intense laser field with both linear and elliptical polarization,

$$\mathbf{E}_{general} = E_0 \begin{pmatrix} \widehat{\mathbf{x}} \alpha \cos\omega t + \widehat{\mathbf{y}} \beta \sin\omega t \\ \widehat{\mathbf{z}} \cos\omega t \end{pmatrix} \tag{5.95}$$

Since the expression is relativistic, we introduce the kinetic energy correction for the Schrödinger equation,. In non-relativistic limit, it is defined as

$$K = \frac{1}{2m}\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right). \tag{5.96}$$

For the relativistic case, we need to do some modification for the kinetic energy term which give us the result as following

$$\begin{aligned} K_{rel} &= \sqrt{\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)^2 c^2 + m^2c^4} - mc^2 \qquad (5.97) \\ &= mc^2\left[\sqrt{1 + \frac{\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)^2}{m^2c^2}} - 1\right] \\ &= mc^2\left\{\sqrt{1 + \left[\frac{1}{mc}\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)\right]^2} - 1\right\} \\ &\approx mc^2\left\{1 + \frac{1}{2}\left[\frac{1}{mc}\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)\right]^2 + \frac{1}{2!}\frac{1}{2}\left(-\frac{1}{2}\right)\left[\frac{1}{mc}\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)\right]^4 + \ldots + -1\right\} \end{aligned}$$

By referring to the above expression, if the second and higher order expansion is removed due to the low velocity limit, we will get back the non-relativistic case which is $\frac{1}{2m}\left[\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)\right]^2$. Next, we make the correction for the relativistic Schrödinger equation of electron at continuum state:

$$\begin{aligned} i\hbar\frac{d}{dt}\Psi_{rel}(\mathbf{r},t) &= mc^2\left\{1 + \frac{1}{2}\left[\frac{1}{mc}\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)\right]^2 + \frac{1}{2!}\frac{1}{2}\left(-\frac{1}{2}\right)\left[\frac{1}{mc}\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)\right]^4 + \ldots + -1\right\} \\ &\quad \times\Psi_{rel}(\mathbf{r},t) \end{aligned} \tag{5.98}$$

Firstly we make an assumption on the wavefunction so that

$$\Psi_{rel}(\mathbf{r},t) = \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) f(t). \tag{5.99}$$

Hence, the Schrödinger equation becomes

$$\begin{aligned} i\hbar\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\frac{\partial}{\partial t}f(t) &= mc^2\{1 + J + K + \ldots + -1\}\Psi_{rel}(\mathbf{r},t) \qquad (5.100) \\ &= mc^2\{1 + J + K + \ldots + -1\}\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) f(t), \qquad (5.101) \end{aligned}$$

with

$$J = \frac{1}{2}\left[\frac{1}{mc}\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)\right]^2, \tag{5.102}$$

$$K = \frac{1}{2!}\frac{1}{2}\left(-\frac{1}{2}\right)\left[\frac{1}{mc}\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)\right]^4. \tag{5.103}$$

In order to make the above eqn E.20 less complicated, let's break it into two major parts

$$\begin{aligned} Y &= \frac{f(t)}{2m}\left[\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)\right]^2 \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) \\ &= \frac{f(t)}{2m}\left[-\hbar^2\nabla^2 - \frac{e\hbar}{i}\nabla\cdot\mathbf{A} - \frac{e\hbar}{i}\mathbf{A}\cdot\nabla + e^2\mathbf{A}^2\right]\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) \end{aligned} \tag{5.104}$$

We recall the coulomb gauge $\nabla\cdot\mathbf{A} = 0$ and yield

$$\nabla\cdot\mathbf{A}\Psi(\mathbf{r},t) = \Psi(\mathbf{x},t)\nabla\cdot\mathbf{A} + \mathbf{A}\cdot\nabla\Psi(\mathbf{r},t) \tag{5.105}$$

$$= \mathbf{A}\cdot\nabla\Psi(\mathbf{r},t), \tag{5.106}$$

imply that the first part

$$\begin{aligned} Y &= \frac{f(t)}{2m}\left[-\hbar^2\nabla^2 - \frac{2e\hbar}{i}\mathbf{A}\cdot\nabla + e^2\mathbf{A}^2\right]\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) \\ &= \frac{f(t)}{2m}\left[p^2 - 2e(\mathbf{A}\cdot\mathbf{p}) + e^2\mathbf{A}^2\right]\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) \\ &= \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\frac{f(t)}{2m}\left[\mathbf{p} - e\mathbf{A}\right]^2, \end{aligned} \tag{5.107}$$

and the second part where

$$\begin{aligned} Z &= \frac{f(t)}{8m^3c^2}\left[\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)\right]^4 \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) \\ &= f(t)\left[(\mathbf{p} - e\mathbf{A})^4\right]\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right), \end{aligned} \tag{5.108}$$

and so on.

Hence, we can rewrite the 5.98 again with this new expression

$$
\begin{aligned}
i\hbar \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\frac{\partial}{\partial t}f(t) &= mc^2 f(t)\{1+J+K+\ldots+-1\}\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right) \\
&= f(t)\left\{\sqrt{(\mathbf{p}-e\mathbf{A})^2c^2+m^2c^4}-mc^2\right\}\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)
\end{aligned} \tag{5.109}
$$

The solution of the time dependent part of the Schrödinger equation can be obtained by solving the differential equation

$$
\int \frac{1}{f(t)}df(t) = -\frac{i}{\hbar}\int\left\{\sqrt{(\mathbf{p}-e\mathbf{A})^2c^2+m^2c^4}-mc^2\right\}dt \tag{5.110}
$$

following by

$$
\ln f(t) = -\frac{i}{\hbar}\left[\int\sqrt{(\mathbf{p}-e\mathbf{A})^2c^2+m^2c^4}dt-\int mc^2dt\right] \tag{5.111}
$$

As a result, the solution obtained is

$$
f(t) = \exp\left\{-\frac{i}{\hbar}\int\left[\sqrt{(\mathbf{p}-e\mathbf{A})^2c^2+m^2c^4}dt-mc^2\right]dt\right\} \tag{5.112}
$$

Then we transform the time-dependent part back and obtain the new relativistic Volkov wavefunction

$$
\begin{aligned}
\Psi_{rel}(\mathbf{r},t) &= \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)f(t) \qquad (5.113)\\
&= \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\exp\left\{-\frac{i}{\hbar}\int\left[\sqrt{(\mathbf{p}-e\mathbf{A})^2c^2+m^2c^4}dt-mc^2\right]dt\right\}
\end{aligned}
$$

By using length gauge transformation, the Volkov wavefunction becomes

$$
\begin{aligned}
\Psi_{\mathrm{P}rel}(\mathbf{r},t) &= \exp\left[-\frac{i}{\hbar}\mathbf{A}(t)\cdot\mathbf{r}\right]\Psi_{rel}(\mathbf{r},t) \qquad (5.114)\\
&= \exp\left[-\frac{i}{\hbar}e\mathbf{A}(t)\cdot\mathbf{r}\right]\exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\exp\left\{-\frac{i}{\hbar}\int_0^t\left[\sqrt{(\mathbf{p}-e\mathbf{A})^2c^2+m^2c^4}-mc^2\right]dt\right\}\\
&= \exp\frac{i}{\hbar}\left\{\Pi(t)\cdot\mathbf{r}-\frac{i}{\hbar}\int_0^t\left[\sqrt{\Pi(t)^2c^2+m^2c^4}-mc^2\right]dt\right\}
\end{aligned}
$$

with $\Pi(t) = \mathbf{p}-e\mathbf{A}(t)$.

5.6 Generalization of Transition Matrix Element

In this case, we set the angular quantum number $l = 0$ and the magnetic quantum number $m = 0$, we may perform the transformation $x = 2r/na_0$ and $b = na_0 B/2$ in the radial integration and obtain

$$V_0 (\Pi(t)) = A \int_0^{\pi} \int_0^{2\pi} F(B) C(t, \phi) \sin^2 \theta d\phi d\theta \tag{5.115}$$

The function $F(B)$ is a b dependent function which is a solution of the integrand $W(r)$

$$\begin{aligned} F(b) &= -\left(\frac{na_0}{2}\right)^3 (-1)^n 2^{3+n} [i(-i+2b)]^{-n-3} (n+1)(n+2) \\ &\quad \times \text{hypergeom}\left([-n, -n+1], [-2-n], \frac{1}{2} i(-i+2b)\right) n \end{aligned} \tag{5.116}$$

The hygeometry function in Eq. 5.116 can be further simplified and a new solution is obtained

$$\begin{aligned} F(b) &= -\left(\frac{na_0}{2}\right)^3 \frac{16in(-1)^n 2^n [-i(i-2b)]^{-n} \left(\frac{1}{2} - ib\right)^{n-1}}{(i-2b)^3 (-1+4ib+4b^2)(-1+2ib)} \\ &\quad \times \left(1 - 2ib - 4in^2 b + 2n^2 - 12b^2 - 12inb + 24ib^3 - 24nb^2\right) \end{aligned} \tag{5.117}$$

After transform back to $B = \left(\frac{2}{na_0}\right) b$, the new expression for $F(B)$ is

$$F(B) = -\left(\frac{na_0}{2}\right)^3 \frac{32n\left(-1+3n^2a^2B^2 - 2n^2 + 6in^2 aB\right)(-1+inaB)^{n-3}}{(1+inaB)^{n+3}} \tag{5.118}$$

The theta integration can be simplified by applying transformation $z = e^{i\theta} = \cos\theta + i\sin\theta$, hence we obtain an semianalytical expression of the transition matrix element

$$V_0 (\Pi(t)) = A \int_0^{2\pi} G(\phi) C(t, \phi) d\phi \tag{5.119}$$

where $G(\phi)$ is the theta integrand of the following expression

$$G(\phi) = \int_0^{\pi} \sin^2\theta F(B)\, d\theta \tag{5.120}$$
$$= Y\int \frac{M(z)}{(z-E)^{n+3}(z-F)^{n+3}}dz$$

with $Y = \frac{i2^3 n}{A^{n+3}}$ and the double poles (from the residue theorem) is obtained

$$E = \frac{-1+\sqrt{1+4AR}}{2A} \tag{5.121}$$
$$F = \frac{-1-\sqrt{1+4AR}}{2A} \tag{5.122}$$

The coeffiecient $A = (Q' + P')$ and $R = (Q' - P')$ are connected to the momentum in x, y and z-plane with the following transformation

$$Q' = \frac{na}{2\hbar}Q \tag{5.123}$$
$$P' = \frac{ina}{2\hbar}P \tag{5.124}$$

Eq. 5.120 can be solved analytically by applying residue theorem and hence the final expression is obtained

$$G(\phi) = 2\pi i \sum_n f(z_n) \tag{5.125}$$
$$= Y\frac{2\pi i}{(n+2)!}\left[\lim_{z\to E}\frac{d^{(n+2)}}{dz^{(n+2)}}\frac{M(z)}{(z-F)^{n+3}} + \lim_{z\to F}\frac{d^{(n+2)}}{dz^{(n+2)}}\frac{M(z)}{(z-E)^{n+3}}\right] \tag{5.126}$$

5.7 Discussions

We have formulated a general model to describe the photoionization of hydrogenic atom via arbitrary initial energy level n, l, m and various intensity of the intense laser source up to the relativistic regime. Firstly, the angular distributions of the differential photoionization rate, $\frac{dw}{d\Theta d\Phi} = \frac{m}{(2\pi\hbar^2)^2}\sum_{k=k_0}^{\infty}|L(\mathbf{p}_k)|^2 p_k \sin\theta$ are plotted by using Eq. (1) at = 0 for linear and circular polarizations with relativistic and nonrelativistic results in Fig. 1. The four scenarios of high and low electric field strength, E and intense laser frequency,

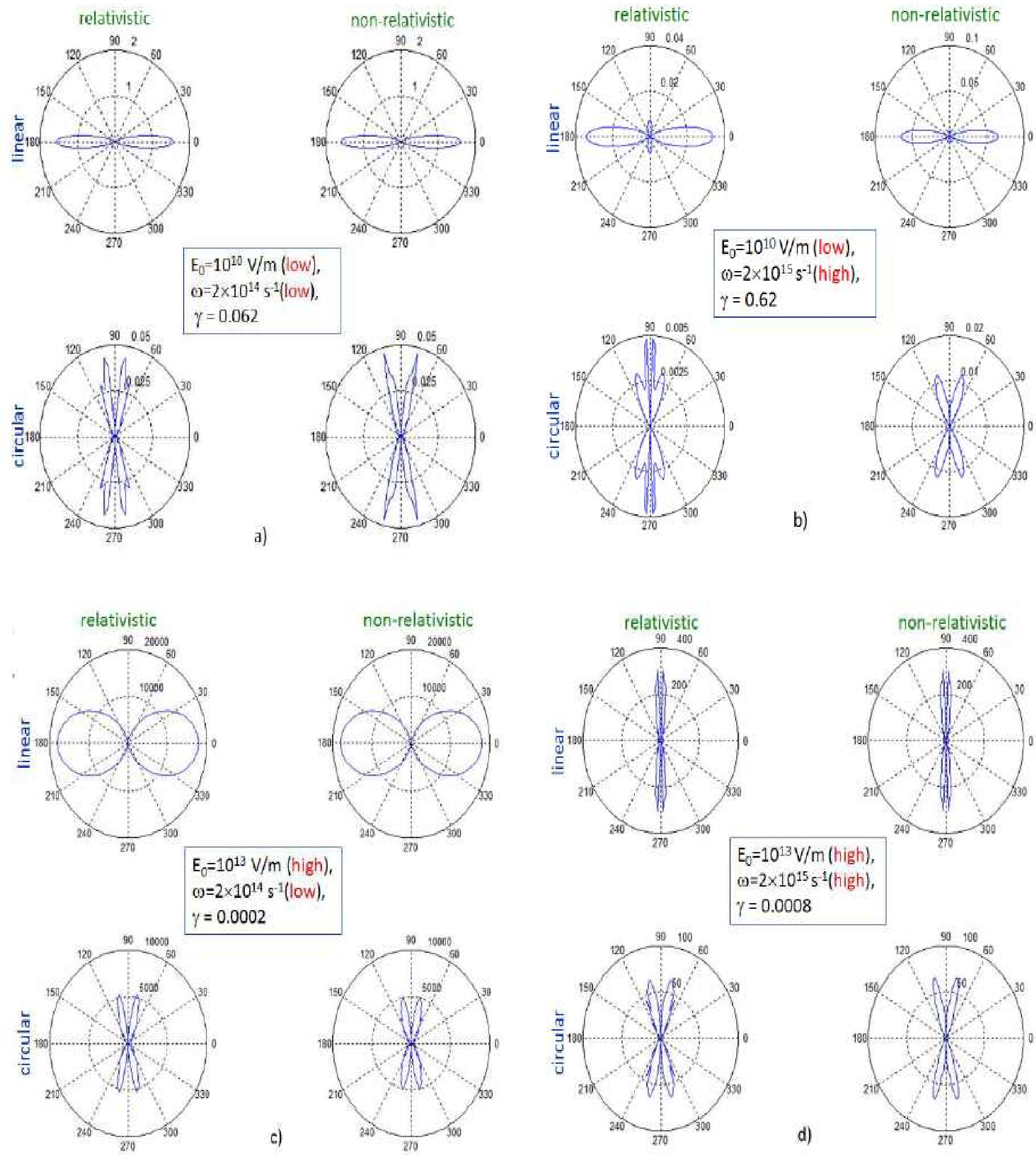


Figure 5.1: (Color online) Angular distributions of photoionization from relativistic (left) and nonrelativistic (right) results for linear and circular polarizations on atom in state $n,l,m = 3,0,0$. The plots are shown for large and small combinations of E_0 and ω.

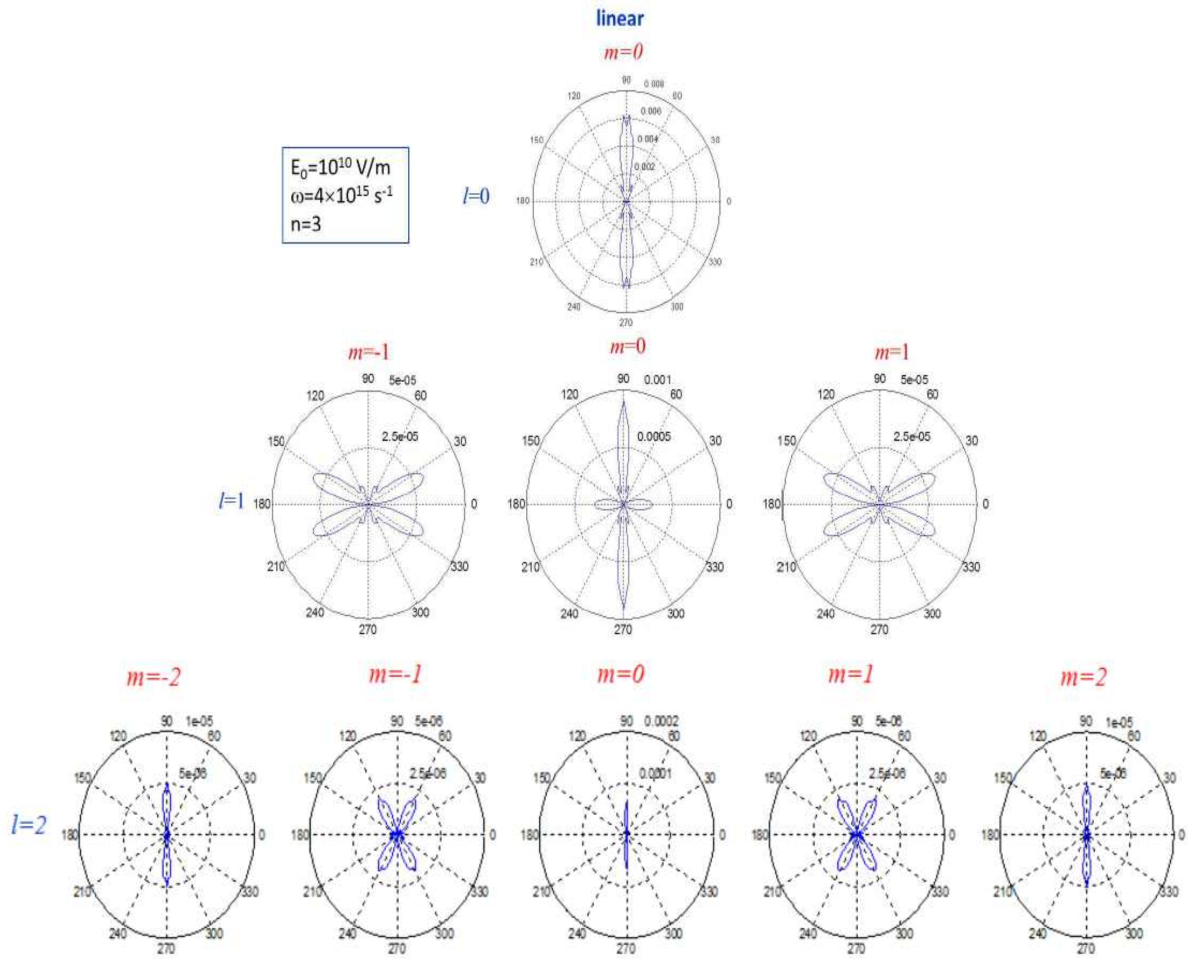


Figure 5.2: (Color online) Angular distribution of photoionization rate for linear polarization with different initial excited atomic states in level $n = 3$.

ω for $n, l, m = 3, 0, 0$ show that the relativistic and non-relativistic results agree very well only for sufficiently small Keldysh parameter $\gamma \approx \frac{\omega}{E}\sqrt{\frac{2mI_p}{e^2}}$, as in case Fig. 1c where the electric field is strong field at low frequency. At high frequency and even with low field, the relativistic effect is significant, as clearly shown in Fig. 1b. This also provides good results on the relativistic photoelectric effect where larger photon energy translates to photoelectron with higher speed. Thus, in the case of larger Keldysh parameter γ, the photoelectron emission probability is much smaller than in the nonrelativistic case and the case of small γ.

Next, we analyze the angular distributions of photoelectron from different orbitals in the excited states. For linear polarized intense laser field as in (Fig. 2), the angular distributions do not depend on the sign of the magnetic quantum number, m. However, for circular polarized(Fig. 3), additional more rounded lobes can be seen for positive m, the emission profiles are non symmetrical against m. In general, the lobes for linear

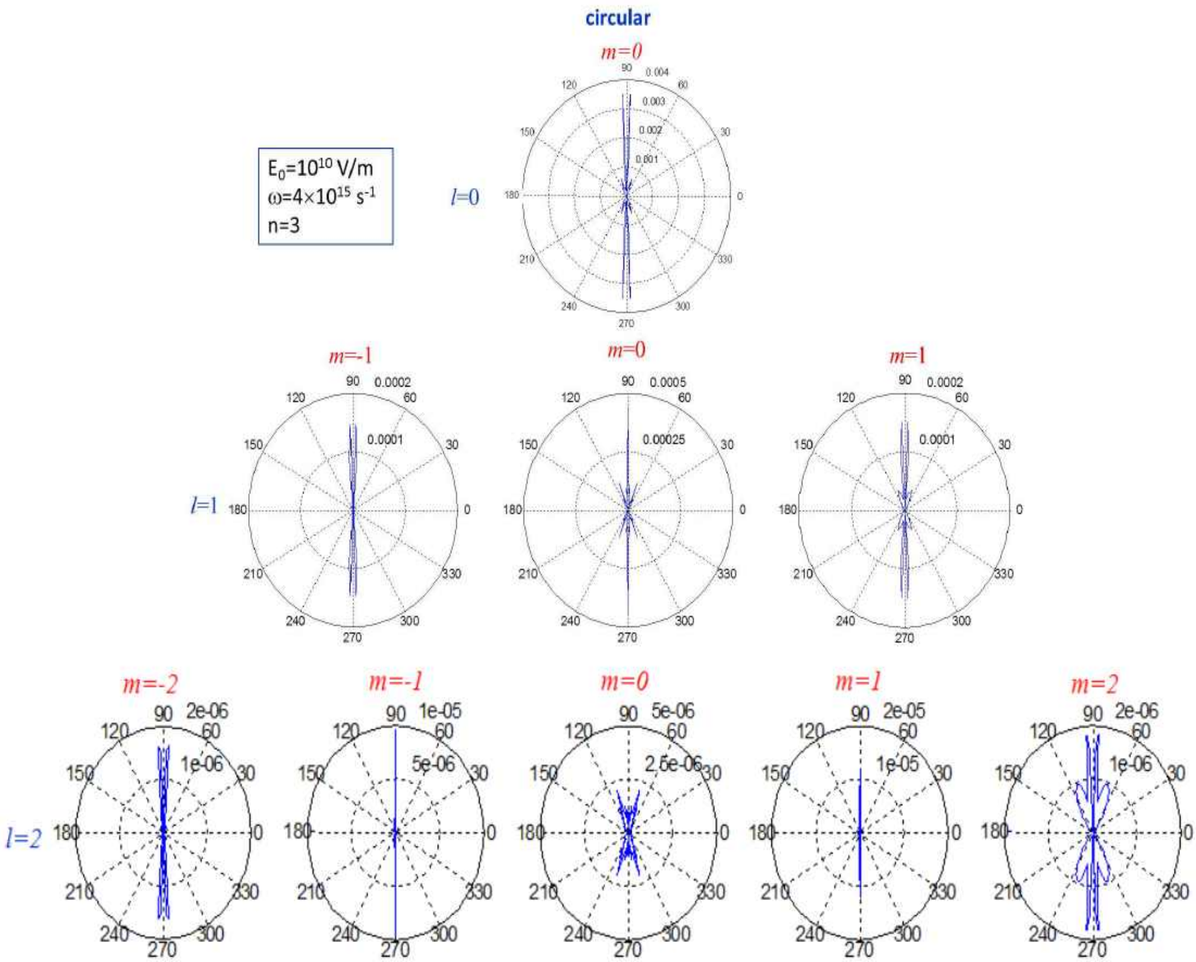


Figure 5.3: (Color online) Angular distribution of photoionization rate for circular polarization with different initial excited atomic states in level $n = 3$.

polarization are almost complementary to the lobes for circular polarization, for instance the minimum in linear case corresponds to the maximum in circular case and vice versa. For linear polarized with $m = 0, \pm 2$ the photoelectron emission rate is the highest mainly at around $\Theta = \pi/2$ and it reduces with l. On the contrary, for the case $m = 1$, there is zero emission towards $\Theta = \pi/2$. This result is counter-intuitive as one would expect that higher excited state would be more likely to be ionized and the electron is ejected predominantly along $\Theta = 0$. For the case of circular polarized intense laser field as in (Fig. 3) with $m = 0$, the are emission is highly directional with twin peaks which are close to $\Theta = \pi/2$.

Subsequently, we look at the angular distributions in excited states for different values of Keldysh parameter, γ . We have plotted the angular distributions of spherically symmetric states $n00$ with $n = 1$ to $n = 4$ in Figs. 4 and 5 for linear and circularpolarizations, respectively. For the case where $\gamma << 1$, multiphoton ionization (MPI) regime,

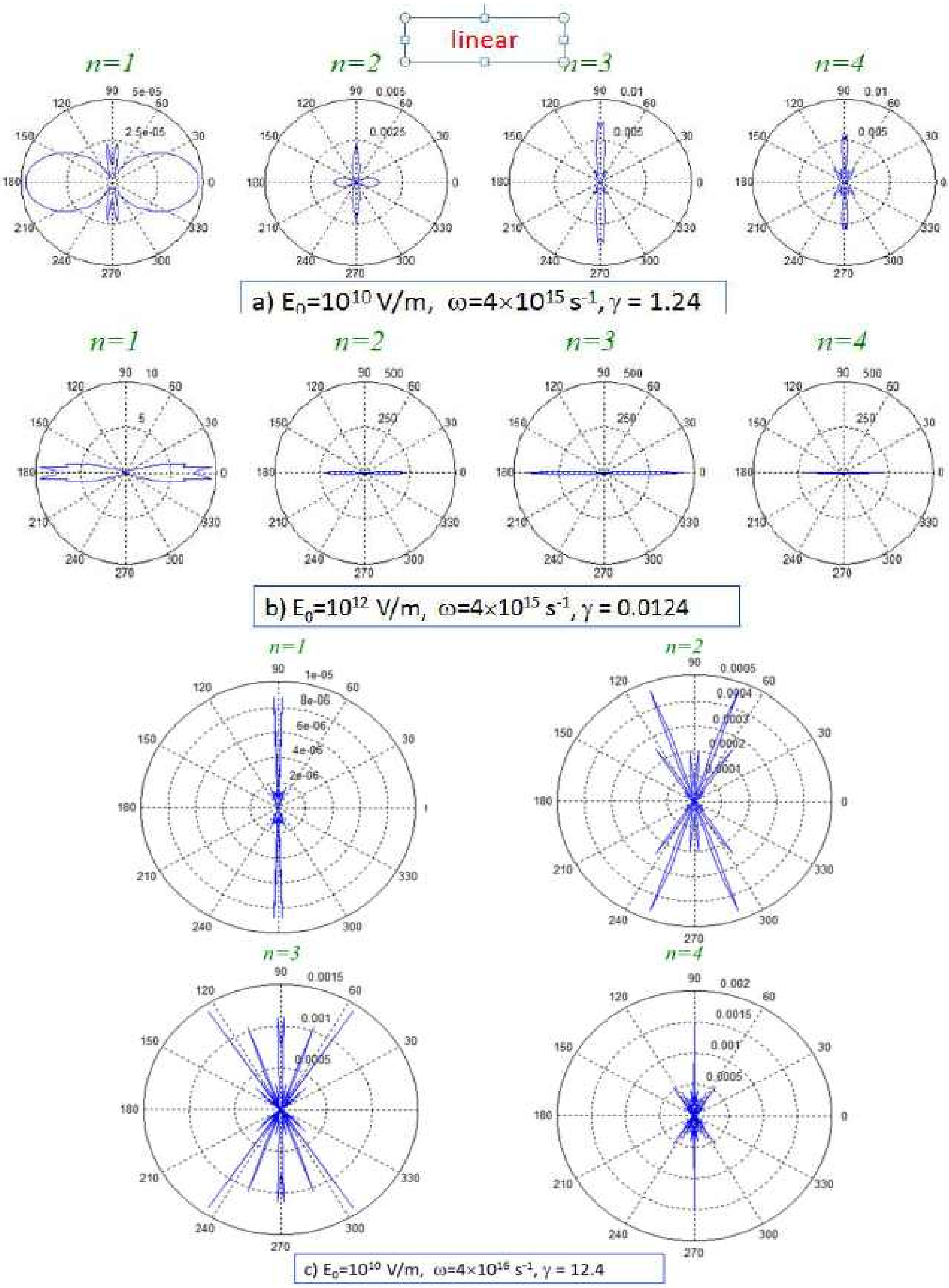


Figure 5.4: (Color online) Angular distributions of photoionization for linear polarization of the first four states $|nlm\rangle = |n00\rangle$ $(n = 1,2\ldots4)$ with: a) $\gamma \sim 1$ b) $\gamma << 1$ c) $\gamma >> 1$.

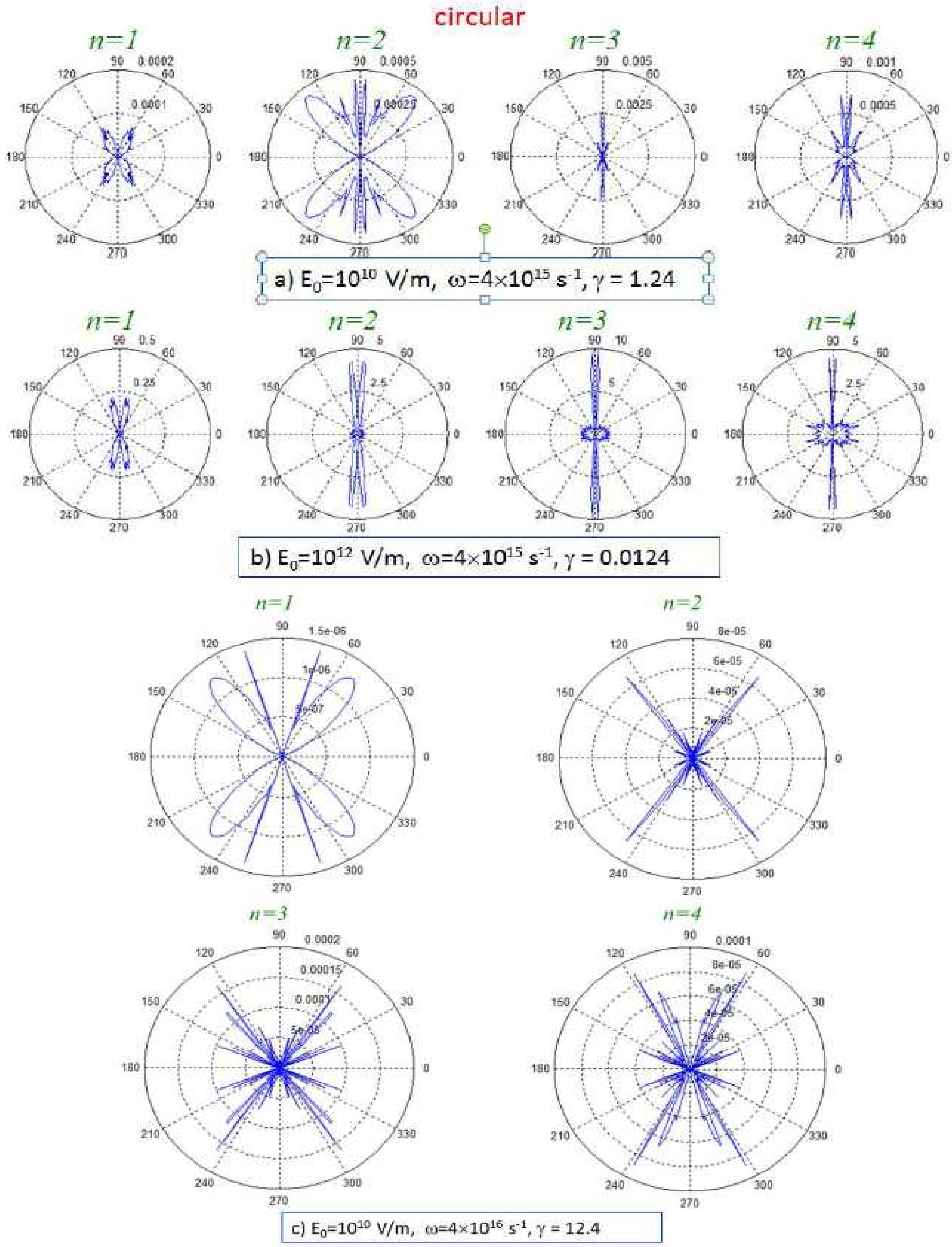


Figure 5.5: (Color online) Angular distributions of photoionization for circular polarization of the first four states $|nlm\rangle = |n00\rangle$ $(n = 1,2\ldots4)$ with: a) $\gamma \sim 1$ b) $\gamma << 1$ c) $\gamma >> 1$.

the photoelectron emission basically follows the direction of the linear polarized electric field, and close to the field direction of the circularly polarized light, especially for higher excited levels where the ionization energies are smaller. Meanwhile for the case where $\gamma \approx 1$, the photoelectron can be emitted into several other directions, especially for lower levels. For $\gamma >> 1$, the tunnel ionization (TI) process plays the dominant, where the photoelectron is emitted into various directions and it becomes hard to distinguish the angular distributions between linear from circular polarized lights. The increased isotropicity in the emission reflects the nature of tunnelling process, which is probabilistic.

We can see that the general trend shown in Figs. 4 and 5 is that the emission rates are typically much larger for linear polarization and the rates increase with the electric field. However, the angular distributions do not change significantly with the electric field strength. The photoionization rate increases with the initial state n up to $n = 3$ and then reduces for larger n. The shape is $\cos\Theta$-like for linear case and $\sin\Theta$-like for circular polarization case to unidirectional, bidirectional close to $\Theta = \pi/2 \pm \varepsilon$, where ε is a small positive value.

In the previous section, we have shown that the relativistic effect on the angular distribution becomes more significant for larger Keldysh parameter γ. In the case of large γ and n, the photoelectrons can be emitted into many discrete directions, with no simple angular distribution. The results also show that photoelectron angular distribution is sensitive to the magnetic quantum number m, which enables us to distinguish the state of a degenerate atom in different internal magnetic states. This could be a useful tool to identify the polarization of the atom by the angular distribution of the photoelectron in the absence of magnetic field, since the different magnetic states cannot be distinguished by spectroscopic data. These results are published in our recent paper (C. H. R. Ooi, Ho, & Bandrauk, 2014).

CHAPTER 6

CONCLUSION AND OUTLOOK

Throughout this research, we have studied the atom-photon interaction and the photoionization of hydrogenic atom in various aspects. It was first learned that the non-perturbative theory main engine to describe the interaction of the atom and intense laser field. Later on, several different interpretations of semi-classical theories which contain different approaches of Hamiltonians were discussed. In particular, the process of light-matter interaction in weak field is studied, where the excitation of an electron due to a less intense laser field, giving a picture of how is the process of electron transition in an atom.

In pursuit of the interaction of atom with weak field, henceforth we have presented several processes on interaction of atom with intense laser field. These phenomenon include above threshold ionization, tunnelling ionization, multiphoton ionization and Corkum's "The Simple Man Model". We discussed the formalism of these processes in details to enable the readers to understand the phenomenon occurred when the intense laser field is playing the major role in the system.

In the second half of this research, we have investigated the process of photoionization through the perturbative approach. We derived the Keldysh's formalism and further extended it to adapt arbitrary momentum of the photoelectron, whereby in Keldysh's model, only the small momentum is considered. Furthermore, we have shown the photoionization spectra of the hydrogenic atom in various fields, for instance linear, circular and elliptical polarized intense laser fields. Later, we made a comparison between our exact model and Keldysh's model. We showed the highly directionality dependence of the photoionization spectra in our model, which provide a very useful information for the experimental setup in detecting the photoionization spectra.

Thus, we have also further generalized our photoionization model by considering arbitrary initial state of the energy level and intense laser field strength. Henceforth, the model has a coverage up to relativistic regime as the electric field strength is increasing.

We have proved the validity of the generalized model agrees with Keldysh's model for certain parameters. Angular distribution of the photoionization spectra is shown and we compare the non-relativistic and relativistic effect on the photoionization spectra. To be surprised, the Keldysh parameter, γ is playing an important role to distinguish the tunnelling ionization regime and multiphoton ionization regime, and hence affecting the directionality of the photoelectron emission.

The generalized theoretical model for photoionization will bring the realization of strong field ionization closer to reality and enhance the field of light matter interaction. The theoretical results would provide useful information and basic prediction as a preparation for experimental verification.

6.1 Research Significances

This research provides a new description of the photoionization spectra in various intense laser fields by using Keldysh-like perturbative approach. The establishment of a complete general model is always a great challenge for a theorist from time to time. Henceforth, this model is very important because it could provide more information for the experimentalist where they meet the bottleneck in experiment due to the lacking of information in the past models.

The generalized model can adapt arbitrary energy level, n,l,m as the initial state of the system. This is very important for the experimental convenience because the initial system is not restricted in the ground state energy level anymore as in the past models. In experiment, the gas sample might be in some excited state after certain process, hence experimentalist can reuse the sample as the initial state of the photoionization system since the theoretical explanation of arbitrary initial energy level is possible. Our model also explain the highly directionality of the photoionization spectra due to various intense laser field. This provide a very good information in experimental setup, so that the detector can be located at the optimized direction to detect the ejected photoelectron.

Furthermore, not to be doubted, the world is concerning on more and more intense laser and higher power facilities such as the Extreme Light Infrastructure (ELI) project. Our research will provide useful physical insight on such project, thus leading to significant knowledge on strong field ionization, in particular enhancing the understanding the connection with the properties of intense light source and the interaction with atom. By using the ultra-intense energy source such as petawatt laser, the relativistic effect is prominent and the photoelectrons are having very high momentum. Our model can explain well for such phenomenon because it adapts arbitrary value of the photoelectron momentum and the relativistic effect is included.

Besides, our research is an important contribution to the future laser system, the attoseconds laser. This laser system is using high harmonic generation as the main driving engine, whereby our model can be further extended into high harmonic generation by taking consideration of the second order perturbation. In spite of that, it provides quite useful information in the excitation process, and plays a good role as the part of the system. In the other word, this research will highly benefit to the intense light-matter interaction and also the development of ultrafast laser.

6.2 Future Works

Given that the advancement of intense laser at a fast pace now, it is expected that the light matter interaction will be a great impediment towards further development. Hence, it is important, both theoretically and experimentally, to understand the photoionization which is a significant process in light matter interaction. With this in mind, the idea of photoionization should be expanded beyond our proof of concept in the paper by (R. Ooi et al., 2012). For instance, a better model should be further developed to include other factors such as the Stark shift effect due to the external electric field. Besides, we are trying to include the magnetic field effect on the photoionization system. The next challenge would be the incorporation of the current model into high harmonic generation. This can be done by taking consideration of the second order perturbation to explain the recollision and recombination process of the photoelectron and its parent ion.

We hope that this idea would transform not only a theoretical framework, but into an experimental verified theory so that it could provide a better physical insight in the future development of intense laser atom interaction.

Appendices

APPENDIX A

LIST OF PUBLICATIONS

JOURNAL ARTICLES

Published

C. H. Raymond Ooi, **WaiLoon Ho**, and A. D. Bandrauk, Photoionization Spectra by Intense Linear, Circular, and Elliptic Polarized Lasers, *Phys. Rev. A* **86**, 023410 (2012). (DOI: 10.1103/PhysRevA.86.023410).

C. H. Raymond Ooi, **W.-L. Ho**, and P. Seow, Orientation Dependent Coherent Anti-Stokes Raman Scattering of Cylindrical Microparticle with Focused Lasers, *J. Opt. Soc. Am. B* **30(9)**, 2427-2435 (2013). (DOI: 10.1364/JOSAB.30.002427).

C. H. Raymond Ooi and **WaiLoon Ho**, Photoelectron Angular Distributions of Excited Atoms in Intense Laser Fields, *Phys. Rev. A* **90**, 013417 (2014). (DOI: 10.1103/PhysRevA.90.013417).

Submitted

WaiLoon Ho, C. H. Raymond Ooi and A. D. Bandrauk, Photoionization of Atoms by Ultrashort Laser Pulses, *Phys. Rev. A*, (2014).

INTERNATIONAL CONFERENCES

WaiLoon Ho & C. H. Raymond Ooi. Generalized Momentum of Tunnelling Ionization of Hydrogenic Atom. Proceeding in IEEE & Oral Presentation in 3rd International Conference on Photonics 2012 (ICP). (2012).

WaiLoon Ho & C. H. Raymond Ooi. Generalization of Photoionization for Hydrogenlike Atom.Oral Presentation in 4th International Meeting on Frontiers of Physics 2013 (IMFP). (2013).

PUBLIC TALK

PhD Seminar

Ho Wai Loon. Orientation Dependent Coherent Anti-Stokes Raman Scattering of Cylindrical Microparticle. PhD Seminar. Auditorium Fizik, Physics Department, University of Malaya. (2012).

APPENDIX B

TIME DEPENDENT PERTURBATION THEORY

It is well known that the time dependent Schrödinger equation is defined by the following expression

$$i\hbar\frac{\partial}{\partial t}|\Xi(t)\rangle = \{H_0 + sV(t)\}|\Xi(t)\rangle. \tag{B.1}$$

In the perturbation ansatz (Langhoff, Epstein, & Karplus, 1972), the wavefunction is written in the form of

$$|\Xi(t)\rangle = \sum_0 s^m |\Xi^{(m)}(t)\rangle, \tag{B.2}$$

where $|\Xi(t)\rangle$ is consisted of all quantum states (Simon, 1973). Hence, the Eq. B.1 becomes

$$i\hbar\sum_0 s^m \frac{\partial}{\partial t}|\Xi^{(m)}(t)\rangle = \sum_0 s^m H_0|\Xi^{(m)}(t)\rangle + \sum_1 s^m V\ (t)|\Xi^{(m-1)}(t)\rangle. \tag{B.3}$$

For $m = 0$, we notice that

$$i\hbar\frac{\partial}{\partial t}|\Xi^{(m)}(t)\rangle = H_0|\Xi^{(0)}(t)\rangle, \tag{B.4}$$

subsequently it gives

$$\sum_n a_n E_n u_n(r)\, e^{-iE_n t/\hbar} = \sum_n a_n H_0 u_n(r)\, e^{-iE_n t/\hbar}. \tag{B.5}$$

As a result, the zeroth order wavefunction takes the form of

$$|\Xi^{(0)}(t)\rangle = \sum_n a_n u_n(r)\, e^{-iE_n t/\hbar}, \tag{B.6}$$

where $H_0 u_n(r) = E_n u_n(r)$ and a_n is a time independent coefficient.

However, for the case where $m \geq 1$, the time dependent Schrödinger equation is

$$i\hbar\frac{\partial}{\partial t}|\Xi^{(m)}(t)\rangle = H_0|\Xi^{(m)}(t)\rangle + V\ (t)|\Xi^{(m-1)}(t)\rangle. \tag{B.7}$$

Generally, we let,

$$\left|\Xi^{(m)}(t)\right\rangle = \sum_n a_n^{(m)}(t)\, u_n(r)\, e^{-iE_n t/\hbar} \tag{B.8}$$

where $a_n^{(m)}(t)$ is the time dependent n-th component coefficient of the m-th order perturbation wavefunction (Stratmann, Scuseria, & Frisch, 1998).

Now, the new expression for Eq. B.7 would be

$$i\hbar \sum_n \frac{\partial a_n^{(m)}(t)}{\partial t} u_n(r)\, e^{-iE_n t/\hbar} = \sum_n a_n^{(m-1)}(t)\, V(t)\, u_n(r)\, e^{-iE_n t/\hbar}, \tag{B.9}$$

and thus it gives us

$$i\hbar \frac{\partial a_n^{(m)}(t)}{\partial t} = \sum_n a_n^{(m-1)}(t)\, V_{kn}(t)\, e^{i(E_k - E_n)t/\hbar} \tag{B.10}$$

$$i\hbar a_k^{(m-1)}(t) = \sum_n \int_{\infty}^{t} a_n^{(m-1)}(t')\, V_{kn}(t')\, e^{i(E_k - E_n)t'/\hbar} dt' \tag{B.11}$$

where the potential of the system is

$$V_{kn}(t) = \langle k|V|n\rangle \tag{B.12}$$

APPENDIX C

THE KRAMERS-HENNEBERGER FRAME

When an atom is placed in a strong field, the Kramers-Henneberger (Reed & Burnett, 1990) is a very important unitary transformation (Sugny et al., 2004) as an approach in order to solve the Schrödinger equation. This transformation is also known as "wiggling" frame (Bhatt, Piraux, & Burnett, 1988). We may start with the minimal coupled time-dependent Schrödinger equation as following

$$\begin{aligned} i\hbar\frac{\partial}{\partial t}\Psi(\mathbf{r},t) &= \left[\frac{1}{2m}\left(\frac{\hbar}{i}\nabla - e\mathbf{A}\right)^2 + V(\mathbf{r})\right]\Psi(\mathbf{r},t) \\ &= \left[-\frac{\hbar^2}{2m}\nabla^2 + \frac{ie\hbar}{m}\mathbf{A}\cdot\nabla + \frac{e^2}{2m}\mathbf{A}^2 + V(\mathbf{r})\right]\Psi(\mathbf{r},t) \end{aligned} \tag{C.1}$$

By introducing two unitary tranformations which are defined as

$$\widehat{U}_1 = \exp\left\{\frac{ie^2}{2m\hbar}\int_{-\infty}^{t} dt'\mathbf{A}^2\right\} \tag{C.2}$$

$$\widehat{U}_2 = \exp\left\{-\frac{e}{m}\int_{-\infty}^{t} dt'\mathbf{A}\cdot\nabla\right\} \tag{C.3}$$

We perform both transformations on the wavefunction according to the following sequence

$$\Psi_{KH}(\mathbf{r},t) = \widehat{U}_2\widehat{U}_1\Psi(\mathbf{r},t) \tag{C.4}$$

where $\Psi_{KH}(\mathbf{r},t)$ is defined as a wavefunction in the Kramers-Henneberger gauge (Grossman, 2008).

Both unitary transformation is very important because the first transformation as in equation C.2 eliminates the squared vector potential. Meanwhile, the second transformation as in equation C.3 locates the coupling into the argument of the potential.

Hence, the new transformed Schrödinger equation in the Kramers-Henneberger frame is shown as below

$$i\hbar\frac{\partial}{\partial t}\Psi_{KH}\left(\mathbf{r},t\right)=\left[-\frac{\hbar^{2}}{2m}\nabla^{2}+V\left[\mathbf{r}+\alpha\left(t\right)\right]\right]\Psi\left(\mathbf{r},t\right) \tag{C.5}$$

where

$$\alpha\left(t\right)=-\frac{e}{m}\int_{-\infty}^{t}dt'\mathbf{A}\left(t'\right) \tag{C.6}$$

APPENDIX D

THE SADDLE POINT METHOD

The saddle point method is used to approximate the asymptotic behavior of integrals, it can be used to approximate $n!$ for large n and also for certain integral such as

$$\int_{-\infty}^{\infty} e^{n\rho(x)} dx \quad \text{and} \quad \int_{C} e^{n\rho(z)} dz \tag{D.1}$$

On the other hand, this method is also known as steepest descent method. Since our purpose is to perform a contour integration on a complex plane, before entering the steepest descent method, we must understand the basic of the complex analysis such as Cauchy Riemann (Folland & Kohn, 1972) condition and analytic function.

D.1 Stirling's approximation

To begin with the saddle point method, firstly we must understand the Stirling's approximation (Kittel & Shore, 1965), where it is used to approximate $n!$ for large n. The Gamma function is defined as below,

$$\Gamma(z) = \int_{0}^{\infty} t^{z-1} e^{-t} dt, \tag{D.2}$$

since $n \in N$, by using integration by parts, let

$$u = t^{n}, \tag{D.3}$$

$$\frac{dv}{dt} = e^{-t}, \tag{D.4}$$

hence, the derivative of the above expression would be

$$\frac{du}{dt} = nt^{n-1}, \tag{D.5}$$

$$v = -e^{-t}. \tag{D.6}$$

Next, we perform the integration by parts to simplify the Gamma function, yielding

$$
\begin{aligned}
\Gamma(n+1) &= \int_0^\infty t^n e^{-t} dt && \text{(D.7)} \\
&= n\Gamma(n) \\
&= n \times (n-1)!\Gamma(1) \\
&= n!. && \text{(D.8)}
\end{aligned}
$$

Then the relation between $n!$ and Gamma function is clearly stated as following

$$
\begin{aligned}
n! &= \Gamma(n+1) && \text{(D.9)} \\
&= \int_0^\infty t^n e^{-t} dt. && \text{(D.10)}
\end{aligned}
$$

By introducing the change of variables $t = nz$ and substitute into the equation D.9, therefore,

$$
\begin{aligned}
n! &= \int_0^\infty (nz)^n e^{-nz} n dz && \text{(D.11)} \\
&= n^{n+1} \int_0^\infty \exp\{n \ln(z)\} e^{-nz} dz && \text{(D.12)} \\
&= n^{n+1} \int_0^\infty \exp(n[\ln(z) - z]) dz. && \text{(D.13)}
\end{aligned}
$$

From the expression above, obviously we can see that $[\ln(z) - z] < 0$ for $z \in (0, \infty)$ and $[\ln(z) - z]$ has maximum at $z = 1$. Hence, $[\ln(z) - z]$ is maximum at $z = 1$ imply that $\exp(n[\ln(z) - z])$ has the maximum value at $z = 1$

As n becomes larger, the difference is becoming more extreme, then the expected dominant contribution is coming from $z = 1$. Therefore, we make a Taylor expansion for the integrand at $z = 1$, letting $g(z) = [\ln(z) - z]$,

$$
\begin{aligned}
n! &= n^{n+1} \int_0^\infty \exp(n[\ln(z) - z]) dz && \text{(D.14)} \\
&\approx n^{n+1} \int_0^\infty \exp\left(n\left[g(1) + \frac{1}{2}(z-1)^2 g''(1)\right]\right) dz && \text{(D.15)} \\
&\approx n^{n+1} e^{-n} \int_0^\infty \exp\left(n\frac{s^2}{2}\right) ds && \text{(D.16)} \\
&\approx n^{n+1} e^{-n} \sqrt{\frac{2\pi}{n}}, && \text{(D.17)}
\end{aligned}
$$

where

$$s = z - 1. \tag{D.18}$$

D.2 Generalization: Steepest Descent Method

Subsequently from the Stirling approximation, next we consider the integrals of the form

$$\int e^{n\rho(x)} dx. \tag{D.19}$$

As the value $n \to \infty$, we have to take an approximation (Battiti, 1992) so that

$$\int_{x_0-\varepsilon}^{x_0+\varepsilon} e^{n\rho(x)} dx \approx \int_R e^{n\rho(x)} dx. \tag{D.20}$$

By the exponential decay of the integrand, Eq. D.20 can be shown that for $a < x_0 < b$,

$$\int_a^b e^{n\rho(x)} dx \ \approx e^{n\rho(x_0)} \sqrt{\frac{2\pi}{n|\rho''(x_0)|}}. \tag{D.21}$$

However, if x_0 is an endpoint, then Eq. D.20 would be

$$\int_a^b e^{n\rho(x)} dx \ \approx e^{n\rho(x_0)} \sqrt{\frac{\pi}{2n|\rho''(x_0)|}}. \tag{D.22}$$

Next, we consider

$$I(n) = \sum_{k=0}^{n} \begin{pmatrix} n \\ k \end{pmatrix} k! n^{-k}, \tag{D.23}$$

and we notice that

$$\int_0^\infty e^{-nx} x^k dx \tag{D.24}$$

Again, we let $t = nx$, $dt = ndx$ so that

$$\int_0^\infty e^{-nx}x^k dx = \int_0^\infty e^{-t}\left(\frac{t}{n}\right)^k n^{-1}dt \quad \text{(D.25)}$$

$$= n^{-k-1}\Gamma(k+1) \quad \text{(D.26)}$$

$$= k!n^{-k-1} \quad \text{(D.27)}$$

Therefore, $I(n)$ can be writen in the summation form,

$$I(n) = \sum_{k=0}^{n}\begin{pmatrix} n \\ k \end{pmatrix} k!n^{-k} \quad \text{(D.28)}$$

$$= \sum_{k=0}^{n}\begin{pmatrix} n \\ k \end{pmatrix} n\int_0^\infty e^{-nx}x^k dx \quad \text{(D.29)}$$

$$= \int_0^\infty e^{-nx}n\left(\sum_{k=0}^{n}\begin{pmatrix} n \\ k \end{pmatrix} x^k\right)dx, \quad \text{(D.30)}$$

henceforth, Eq. D.28 can be further reduced as the following expression

$$I(n) = \int_0^\infty e^{-nx}n(1+x)^n dx \quad \text{(D.31)}$$

$$= n\int_0^\infty \exp(n[\ln(1+x)-x])\,dx. \quad \text{(D.32)}$$

Now, we take $\rho(x) = \ln(1+x) - x$ and imply that

$$\rho'(x) = \frac{1}{(1+x)} - 1, \quad \text{(D.33)}$$

such that $x_0 = 0$ (an endpoint).

$$\rho(0) = \ln(1) - 0 \quad \text{(D.34)}$$

$$= 0, \quad \text{(D.35)}$$

and the second derivative would be

$$|\rho''(0)| = \left|\frac{-1}{(1+0)^2}\right| \quad \text{(D.36)}$$

$$= |-1| \quad \text{(D.37)}$$

$$= 1. \quad \text{(D.38)}$$

Hence, by using Laplace's method, we obtain the value for I_n

$$
\begin{aligned}
I(n) &= \sum_{k=0}^{n} \begin{pmatrix} n \\ k \end{pmatrix} k! n^{-k} && \text{(D.39)}\\
&\approx e^{n\rho(x_0)} \sqrt{\frac{\pi}{2n|\rho''(x_0)|}} && \text{(D.40)}\\
&= \sqrt{\frac{n\pi}{2}}. && \text{(D.41)}
\end{aligned}
$$

Consequently, we extend the formalism onto the complex plane so that

$$\int_C e^{n\rho(z)} dz = \int_C e^{nRe\rho(z)} e^{niIm\rho(z)} dz, \tag{D.42}$$

where $\rho(z)$ is an analytic function on C.

The strategy for $Re\rho$ is analogous to the real case as previous note. The main importance here is to solve the $Im\rho$ part. Furthermore, the main idea for this is to deform the contour C to C' so that $Im\rho$ is a constant on C'. Then,

$$\int_C e^{n\rho(z)} dz = e^{niIm\rho(z)} \int_{C'} e^{nRe\rho(z)} dz \tag{D.43}$$

After getting this formation, Laplace method is applied to find C',

$$
\begin{aligned}
\rho(z) &= u(x,y) + iv(x,y) && \text{(D.44)}\\
&= u(x,y) + iv(x_0,y_0) && \text{(D.45)}
\end{aligned}
$$

where $z_0 = x_0 + iy_0$ and $\rho'(z_0) = 0$. (The dominant contribution is contributed by z_0.

Let us recall from the expression from Steepest descent, for $v(x,y) = v(x_0,y_0)$,

$$
\begin{aligned}
\nabla v &= \left(\frac{\partial v}{\partial x}, \frac{\partial v}{\partial y}\right) && \text{(D.46)}\\
&= \left(-\frac{\partial u}{\partial y}, \frac{\partial u}{\partial x}\right) && \text{(D.47)}
\end{aligned}
$$

The direction tangent to this curve is

$$\left(\frac{\partial u}{\partial x}, \frac{\partial u}{\partial y}\right) = \nabla u, \tag{D.48}$$

where this is the steepest descent in u. Next, we consider the asymptotics of

$$\frac{1}{\Gamma(z)} = \frac{1}{2\pi i}\int_C t^{-z}e^t\,dt. \tag{D.49}$$

The expression is similar to the Hankel contour (Arfken & Weber, 2005).

$$\begin{aligned}
\frac{1}{\Gamma(z)} &= \frac{1}{2\pi i}\int_C t^{-z}e^t\,dt && \text{(D.50)}\\
&= \frac{1}{2\pi i}\int_C \exp\left(\ln t^{-z}\right)e^t\,dt && \text{(D.51)}\\
&= \frac{1}{2\pi i}\int_C \exp\left[t - z(\ln t)\right]dt. && \text{(D.52)}
\end{aligned}$$

Again, we let $t = sz$, $dt = zds$, and yield

$$\begin{aligned}
\frac{1}{\Gamma(z)} &= \frac{1}{2\pi i}z\int_C \exp\left[sz - z(\ln sz)\right]ds && \text{(D.53)}\\
&= \frac{1}{2\pi i}zz^{-z}\int_C \exp\left[sz - z\ln s\right]ds && \text{(D.54)}\\
&= \frac{1}{2\pi i z^{z-1}}\int_C \exp\left[z(s-\ln s)\right]ds. && \text{(D.55)}
\end{aligned}$$

Now, we take $\rho(s) = (s - \ln s)$ and the derivative would be

$$\rho'(s) = 1 - \frac{1}{s}, \tag{D.56}$$

such that $\rho'(1) = 0$

If we deform C to pass through $z = 1$ and hold $Im\rho(z)$ constant then the dominant contribution will be around $z = 1$.In order to hold $Im\rho(z)$ as a constant, henceforth, we set the variable $\rho(z)$ only in the imaginary direction. We let $z = 1 + iv$, then

$$\begin{aligned}
\rho(v) &= 1 + iv - \ln(1+iv) && \text{(D.57)}\\
&= 1 - \frac{v^2}{2} + \frac{iv^3}{3} + \ldots && \text{(D.58)}\\
&\approx 1 - \frac{v^2}{2} && \text{(D.59)}
\end{aligned}$$

Next, by letting $g(v) = 1 - \frac{v^2}{2}$, we have

$$\frac{1}{\Gamma(z)} = \frac{1}{2\pi i z^{z-1}}\int_C \exp\left[z(s-\ln s)\right]ds \tag{D.60}$$

$$\approx \frac{1}{2\pi i z^{z-1}}\int_{-\varepsilon}^{\varepsilon} \exp\left[zg(v)\right]dv \tag{D.61}$$

$$\approx \frac{1}{2\pi i z^{z-1}}e^{z}\int_{-\infty}^{\infty} \exp\left[\left(-z\frac{v^2}{2}\right)\right]dv, \tag{D.62}$$

and

$$\frac{1}{\Gamma(z)} = \frac{1}{2\pi z^{z-1}}e^{z}\sqrt{\frac{2\pi}{z}} \tag{D.63}$$

$$= \left(\frac{e}{z}\right)^{z}\sqrt{\frac{z}{2\pi}}, \tag{D.64}$$

where $|\rho''(z_0)| = 1$.

In generalization, it can be shown that to leading order:

$$\int_C e^{n\rho(z)}dz \approx e^{i\theta}e^{n\rho(z_0)}\sqrt{\frac{\pi}{k|\rho''(z_0)|}} \tag{D.65}$$

where

$$\theta = -\frac{\alpha}{2}+\frac{\pi}{2}, -\frac{\alpha}{2}+\frac{3\pi}{2}, \tag{D.66}$$

is the direction of steepest descent, and

$$|\rho''(z_0) = |\rho''(z_0)|e^{-i\alpha} \tag{D.67}$$

APPENDIX E

MOMENTUM SPACE WAVEFUNCTION TRANSFORMATION

The general equation for hydrogen ground state wavefunction is defined as the following

$$\begin{aligned}\Psi_{n,l,m}(\mathbf{r},\theta,\phi) &= \left\{\frac{1}{(2\pi)^{1/2}}e^{\pm im\phi}\right\}\left\{\left(\frac{(2l+1)(l-m)!}{2(l+m)!}\right)^{\frac{1}{2}}P_l^m(\cos\theta)\right\} \\ &\times\left\{\frac{(2Y)^{l+1}}{(n+l)!}\left(\frac{Y(n-l-1)!}{n(n+l)!}\right)^{\frac{1}{2}}\exp(-Yr)\,r^l L_{n+l}^{2l+1}(2Yr)\right\},\end{aligned} \tag{E.1}$$

where the definition for $Y = \frac{Z}{na_0}$.

$P_l^m(\cos\Theta)$ is called Ferrers' associated Legendre function and $L_{n+l}^{2l+1}(2Yr)$ is called associated Laguarre polynomial which is defined by the identity

$$\sum_{\beta=0}^{\infty}\frac{L_{\alpha+\beta}^{\alpha}(\xi)}{(\alpha+\beta)!}u^{\beta} = (-1)^{\alpha}\frac{\exp\left(-\frac{\xi u}{1-u}\right)}{(1-u)^{\alpha+1}}. \tag{E.2}$$

Next, we define the direction vector $\mathbf{x},\mathbf{y}$ and $\mathbf{z}$ as

$$\mathbf{x} = r\sin\theta\cos\phi\widehat{\mathbf{x}}, \tag{E.3}$$

$$\mathbf{y} = r\sin\theta\sin\phi\widehat{\mathbf{y}}, \tag{E.4}$$

$$\mathbf{z} = r\cos\theta\widehat{\mathbf{z}}, \tag{E.5}$$

and the momentum vector according to each direction

$$\mathbf{p}_x = P\sin\Theta\cos\Phi\widehat{\mathbf{x}}, \tag{E.6}$$

$$\mathbf{p}_y = P\sin\Theta\sin\Phi\widehat{\mathbf{y}}, \tag{E.7}$$

$$\mathbf{p}_z = P\cos\Theta\widehat{\mathbf{z}}, \tag{E.8}$$

where the magnitude of the momentum is

$$P = \sqrt{p_x^2+p_y^2+p_z^2}. \tag{E.9}$$

Subsequently, the dot product between the momentum and the direction vector would be

$$\begin{aligned}
\mathbf{P}\cdot\mathbf{r} &= rP\left[\sin\Theta\cos\Phi\sin\theta\cos\phi\,(\widehat{\mathbf{x}}\cdot\widehat{\mathbf{x}})+\sin\Theta\sin\Phi\sin\theta\sin\phi\,(\widehat{\mathbf{y}}\cdot\widehat{\mathbf{y}})+\cos\Theta\cos\theta\,(\widehat{\mathbf{z}}\cdot\widehat{\mathbf{z}})\right] \\
&= rP\left[\sin\Theta\sin\theta\,(\cos\Phi\cos\phi+\sin\Phi\sin\phi)+\cos\Theta\cos\theta\right] \\
&= rP\left[\sin\Theta\sin\theta\cos(\Phi-\phi)+\cos\Theta\cos\theta\right] \qquad \text{(E.10)}
\end{aligned}$$

By using the trigonometry identities

$$\cos(\Phi\mp\phi)=\cos\Phi\cos\phi\pm\sin\Phi\sin\phi, \qquad \text{(E.11)}$$

then the transformation of momentum eigenfunction is given by

$$\begin{aligned}
\Psi_{n,l,m}(P,\Theta,\Phi) &= \int\exp\left[-\frac{i}{\hbar}\mathbf{P}\cdot\mathbf{r}\right]\times\Psi_{n,l,m}(\mathbf{r},\theta,\phi)\,d^3r \qquad \text{(E.12)} \\
&= \int_0^\infty\int_0^\pi\int_0^{2\pi}\Psi_{n,l,m}(\mathbf{r},\theta,\phi) \qquad \text{(E.13)} \\
&\quad\times\exp\left[-\frac{i}{\hbar}(\sin\Theta\sin\theta\cos(\Phi-\phi)+\cos\Theta\cos\theta)\,rP\right]r^2\sin\theta\,drd\theta d\phi \\
&= \int_0^\infty\int_0^\pi\int_0^{2\pi}LMNr^2\sin\theta\,drd\theta d\phi, \qquad \text{(E.14)}
\end{aligned}$$

with

$$\begin{aligned}
L &= \left\{\frac{1}{(2\pi)^{1/2}}e^{\pm im\phi}\right\}\left\{\left(\frac{(2l+1)(l-m)!}{2(l+m)!}\right)^{\frac{1}{2}}P_l^m(\cos\theta)\right\}, \qquad \text{(E.15)} \\
M &= \left\{\frac{(2Y)^{l+1}}{(n+l)!}\left(\frac{Y(n-l-1)!}{n(n+l)!}\right)^{\frac{1}{2}}\exp(-Yr)\,r^lL_{n+l}^{2l+1}(2Yr)\right\}, \qquad \text{(E.16)} \\
N &= \exp\left[-\frac{i}{\hbar}(\sin\Theta\sin\theta\cos(\Phi-\phi)+\cos\Theta\cos\theta)\,rP\right]. \qquad \text{(E.17)}
\end{aligned}$$

We introduce an important transformation which is

$$\begin{aligned}
I_1 &= \int_0^{2\pi}\exp\left[\pm im\phi+ib\cos(\Phi-\phi)\right]d\phi, \qquad \text{(E.18)} \\
b &= -\frac{rP}{\hbar}\sin\theta\sin\Theta, \qquad \text{(E.19)}
\end{aligned}$$

and

$$
\begin{aligned}
I_2 &= \int_0^{\pi} I_1 P_l^m(\cos\theta)\sin\theta\exp\left[id\cos\theta\cos\Theta\right]d\theta, \\
d &= -\frac{rP}{\hbar}.
\end{aligned}
$$

Therefore, let us rearrange the momentum eigenfunction after introducing the replacing I_1 and I_2 into Eq. E.14

$$
\begin{aligned}
\Psi_{n,l,m}(P,\Theta,\Phi) &= \frac{1}{(2\pi)^{1/2}}\left(\frac{(2l+1)(l-m)!}{2(l+m)!}\right)^{\frac{1}{2}}\frac{(2Y)^{l+1}}{(n+l)!}\left(\frac{Y(n-l-1)!}{n(n+l)!}\right)^{\frac{1}{2}} \qquad \text{(E.20)} \\
&\times\int_0^{\infty} I_2\exp(-Yr)\,r^{l+2}L_{n+l}^{2l+1}(2Yr)\,dr
\end{aligned}
$$

Next, we evaluate the I_1 by introducing a new transformation $\phi-\Phi=w$, hence

$$
\begin{aligned}
I_1 &= \int_0^{2\pi}\exp\left[\pm im\phi+ib\cos(\Phi-\phi)\right]d\phi && \text{(E.21)} \\
&= \int_0^{2\pi}\exp\left[\pm im(w+\Phi)+ib\cos w\right]dw && \text{(E.22)} \\
&= e^{\pm im\Phi}2\pi i^{\pm m}J_{\pm m}(b) && \text{(E.23)}
\end{aligned}
$$

The Sommerfield's integral gives the solution of Bessel function of order $\pm m$. Note that this expression

$$
J_{-m}(b)=i^{2m}J_m(b), \qquad \text{(E.24)}
$$

give us the negative m solution

$$
\begin{aligned}
I_1 &= e^{-im\Phi}2\pi i^{-m}J_{-m}(b) && \text{(E.25)} \\
&= e^{-im\Phi}2\pi i^{-m}i^{2m}J_m(b) && \text{(E.26)} \\
&= e^{-im\Phi}2\pi i^{m}J_m(b). && \text{(E.27)}
\end{aligned}
$$

Consequently, we can generalize the solution for the integral I_1 as

$$
I_1=2\pi i^m J_m(b)\,e^{\pm im\Phi} \qquad \text{(E.28)}
$$

where

$$
\begin{aligned}
b &= d \sin\theta \sin\Theta && \text{(E.29)} \\
d &= -\frac{rP}{\hbar} && \text{(E.30)}
\end{aligned}
$$

Next, our upcoming task is to evaluate the I_2 integral. Firstly, let us define the generating function of the Gegenbauer's polynomial

$$
\begin{aligned}
Q_\nu &\equiv \frac{1}{\left(1+ut+u^2\right)^\nu} && \text{(E.31)} \\
&\equiv \sum_{k=0}^{\infty} C_k^\nu(t)\, u^k && \text{(E.32)}
\end{aligned}
$$

When we set $\nu = \frac{1}{2}$, then the function reduce to the Legendre polynomials. By putting $\nu = \frac{1}{2}$ and differentiate the function m times with respect of t, then we obtain

$$P_l^m(t) = 1\cdot 3\cdot 5\cdots(2m-1)\left(1-t^2\right)^{m/2} C_{l-m}^{m+1/2}(t) \qquad \text{(E.33)}$$

In 1877, Gegenbauer evaluate the following integral

$$
\begin{aligned}
&\int_0^\pi e^{iz\cos\theta\cos\psi} J_{\nu-1/2}(z\sin\theta\sin\psi)\, C_r^\nu(\cos\theta)\sin^{\nu+1/2}\theta\, d\theta && \text{(E.34)} \\
&= \left(\frac{2\pi}{z}\right)^{1/2} i^r \left(\sin^{\nu-1/2}\psi\right) C_r^\nu(\cos\psi)\, J_{\nu+r}(z)
\end{aligned}
$$

By multiplying a factor of $1\cdot 3\cdot 5\cdots(2m-1)\left(1-t^2\right)^{m/2}$ and putting $\nu = m+1/2$; $z = d$; $x = \Theta$ and $r = l-m$

$$
\begin{aligned}
&1\cdot 3\cdot 5\cdots(2m-1)\left(1-t^2\right)^{m/2} \int_0^\pi e^{id\cos\theta\cos\Theta} J_m(d\sin\theta\sin\Theta)\, C_{l-m}^{m+1/2}(\cos\theta)\sin^{m+1}\theta\, d\theta \\
&= 1\cdot 3\cdot 5\cdots(2m-1)\left(1-t^2\right)^{m/2} \left(\frac{2\pi}{d}\right)^{1/2} i^{l-m}\left(\sin^0\psi\right) C_{l-m}^{m+1/2}(\cos\Theta)\, J_{l+1/2}(d) && \text{(E.35)} \\
&= \left(\frac{2\pi}{d}\right)^{1/2} i^{l-m} P_l^m(\cos\Theta)\, J_{l+1/2}(d) && \text{(E.36)}
\end{aligned}
$$

With the aid of equation E.34, we can solve for the integral I_2, yielding

$$
\begin{aligned}
I_2 &= \int_0^{\pi} I_1 P_l^m(\cos\theta)\sin\theta \exp[id\cos\theta\cos\Theta]\,d\theta && \text{(E.37)}\\
&= \int_0^{\pi} 2\pi i^m J_m(b)\, e^{\pm im\Phi} P_l^m(\cos\theta)\sin\theta\exp[id\cos\theta\cos\Theta]\,d\theta && \text{(E.38)}\\
&= 2\pi i^m e^{\pm im\Phi}\int_0^{\pi}\exp[id\cos\theta\cos\Theta]\,J_m(d\sin\theta\sin\Theta)\,P_l^m(\cos\theta)\sin\theta d\theta && \text{(E.39)}
\end{aligned}
$$

By reducing I_2 into $2\pi i^m e^{\pm im\Phi}\left[1\cdot 3\cdot 5\cdots(2m-1)\left(1-t^2\right)^{m/2}\right]$

$\times\int_0^{\pi}\exp[id\cos\theta\cos\Theta]\,J_m(d\sin\theta\sin\Theta)\,C_{l-m}^{m+1/2}(\cos\theta)\sin\theta d\theta$, then we obtain the solution for I_2, which is

$$
\begin{aligned}
I_2 &= 2\pi i^m e^{\pm im\Phi}\left(\frac{2\pi}{d}\right)^{1/2} i^{l-m} P_l^m(\cos\Theta)\,J_{l+1/2}(d) && \text{(E.40)}\\
&= 2\pi i^l e^{\pm im\Phi}\left(-\frac{2\pi\hbar}{rP}\right)^{1/2} P_l^m(\cos\Theta)\,J_{l+1/2}\left(-\frac{rP}{\hbar}\right) && \text{(E.41)}\\
&= -2\pi(-i)^l e^{\pm im\Phi}\left(\frac{2\pi\hbar}{P}\right)^{1/2} r^{-1/2} P_l^m(\cos\Theta)\,J_{l+1/2}\left(\frac{rP}{\hbar}\right) && \text{(E.42)}
\end{aligned}
$$

After that, the final task would be the solution of the radial integral. We refer to equation E.20, the main part for the radial integral

$$
\begin{aligned}
\Psi_{n,l,m}(P,\Theta,\Phi) &= A\int_0^{\infty} I_2\exp(-Yr)\,r^{l+2}L_{n+l}^{2l+1}(2Yr)\,dr && \text{(E.43)}\\
&= A\int_0^{\infty} r^{-1/2}J_{l+1/2}\left(\frac{rP}{\hbar}\right)\exp(-Yr)\,r^{l+2}L_{n+l}^{2l+1}(2Yr)\,dr, && \text{(E.44)}
\end{aligned}
$$

where A is the coeficient

$$
\begin{aligned}
A &= \frac{1}{(2\pi)^{1/2}}\left(\frac{(2l+1)(l-m)!}{2(l+m)!}\right)^{\frac{1}{2}}\frac{(2Y)^{l+1}}{(n+l)!}\left(\frac{Y(n-l-1)!}{n(n+l)!}\right)^{\frac{1}{2}} && \text{(E.45)}\\
&\quad\left\{-2\pi(-i)^l e^{\pm im\Phi}\left(\frac{2\pi\hbar}{P}\right)^{1/2} P_l^m(\cos\Theta)\right\}.
\end{aligned}
$$

We focus on the main part of the radial integral that contains

$$
\int_0^{\infty} r^{l+3/2}J_{l+1/2}\left(\frac{rP}{\hbar}\right)\exp(-Yr)\,L_{n+l}^{2l+1}(2Yr)\,dr. \qquad \text{(E.46)}
$$

Then, we introduce a new transformation by substitution

$$\begin{aligned} \eta &= 2Yr & \text{(E.47)} \\ \zeta &= \frac{P}{Y\hbar} & \text{(E.48)} \end{aligned}$$

and

$$\frac{d\eta}{dr} = 2Y \qquad \text{(E.49)}$$

Henceforth, we have

$$\begin{aligned} & \int_0^\infty r^{l+3/2} J_{l+1/2}\left(\frac{rP}{\hbar}\right) \exp\left(-Yr\right) L_{n+l}^{2l+1}\left(2Yr\right) dr \qquad \text{(E.50)} \\ = \; & (2Y)^{-(l+5/2)} \int_0^\infty \eta^{l+3/2} J_{l+1/2}\left(\frac{1}{2}\zeta\eta\right) \exp\left(-\frac{\eta}{2}\right) L_{n+l}^{2l+1}\left(\eta\right) d\eta. \end{aligned}$$

In order simplify the above expression, let us define the η integral by

$$I_{nl}\left(\zeta\right) = \int_0^\infty \exp\left(-\frac{\eta}{2}\right) \eta^{l+3/2} J_{l+1/2}\left(\frac{1}{2}\zeta\eta\right) L_{n+l}^{2l+1}\left(\eta\right) d\eta. \qquad \text{(E.51)}$$

By introducing a new function U with the following identity

$$\begin{aligned} U &\equiv U_l\left(\zeta, u\right) & \text{(E.52)} \\ &\equiv \sum_{n=l+1}^{\infty} \frac{I_{nl}\left(\zeta\right)}{(n+l)!} u^{n-l-1} & \text{(E.53)} \end{aligned}$$

Then we evaluate the function by using the generating function for the associated Laguarre polynomials and thus obtaining $I_{nl}\left(\zeta\right)$ as coeffiecients of th expansion of $U_l\left(\zeta, u\right)$ as a power series in u

$$\begin{aligned} U &= \sum_{n=l+1}^{\infty} \frac{I_{nl}\left(\zeta\right)}{(n+l)!} u^{n-l-1} & \text{(E.54)} \\ &= \int_0^\infty \sum_{n=l+1}^{\infty} \frac{\exp\left(-\frac{\eta}{2}\right) \eta^{l+3/2} J_{l+1/2}\left(\frac{1}{2}\zeta\eta\right) L_{n+l}^{2l+1}\left(\eta\right)}{(n+l)!} u^{n-l-1} d\eta & \text{(E.55)} \\ &= \int_0^\infty \exp\left(-\frac{\eta}{2}\right) \eta^{l+3/2} J_{l+1/2}\left(\frac{1}{2}\zeta\eta\right) \sum_{n=l+1}^{\infty} \frac{L_{n+l}^{2l+1}\left(\eta\right)}{(n+l)!} u^{n-l-1} d\eta & \text{(E.56)} \end{aligned}$$

Since we know that the identity as following

$$\sum_{\beta=0}^{\infty}\frac{L_{\alpha+\beta}^{\alpha}(\xi)}{(\alpha+\beta)!}u^{\beta}=(-1)^{\alpha}\frac{\exp\left(-\frac{\xi u}{1-u}\right)}{(1-u)^{\alpha+1}}, \tag{E.57}$$

then we have

$$\sum_{n=l+1}^{\infty}\frac{L_{n+l}^{2l+1}(\eta)}{(n+l)!}u^{n-l-1}=(-1)^{2l+1}\frac{\exp\left(-\frac{\eta u}{1-u}\right)}{(1-u)^{2l+2}}. \tag{E.58}$$

We substitute Eq. E.58 into Eq. E.56 and yield

$$\begin{aligned}
U &= \int_0^{\infty}\exp\left(-\frac{\eta}{2}\right)\eta^{l+3/2}J_{l+1/2}\left(\frac{1}{2}\zeta\eta\right)\sum_{n=l+1}^{\infty}\frac{L_{n+l}^{2l+1}(\eta)}{(n+l)!}u^{n-l-1}d\eta && \text{(E.59)}\\
&= \int_0^{\infty}\exp\left(-\frac{\eta}{2}\right)\eta^{l+3/2}J_{l+1/2}\left(\frac{1}{2}\zeta\eta\right)(-1)^{2l+1}\frac{\exp\left(-\frac{\eta u}{1-u}\right)}{(1-u)^{2l+2}}d\eta && \text{(E.60)}\\
&= \frac{(-1)^{2l+1}}{(1-u)^{(2l+2)}}\int_0^{\infty}\eta^{l+3/2}J_{l+1/2}\left(\frac{1}{2}\zeta\eta\right)\exp\left[-\eta\frac{1+u}{2(1-u)}\right]d\eta. && \text{(E.61)}
\end{aligned}$$

The integral in Eq. E.61 had been evaluated by Hankel and Gegenbauer once upon a time, and it can be declared as an identity here

$$\int_0^{\infty}x^{\mu-1}J_{\nu}(zx)\exp[-\alpha x]\,dx=\frac{(z/2\alpha)^{\nu}\,\Gamma(\mu+\nu)}{\alpha^{\mu+\nu}\Gamma(\nu+1)}F\left(\frac{\mu+\nu}{2},\frac{\mu+\nu+1}{2};\nu+1;-\frac{z^2}{\alpha^2}\right). \tag{E.62}$$

Again, we perform a transformation and let

$$\begin{aligned}
z &= \frac{1}{2}\zeta, && \text{(E.63)}\\
\nu &= l+\frac{1}{2}, && \text{(E.64)}\\
\mu &= l+\frac{5}{2}, && \text{(E.65)}\\
\alpha &= \frac{1}{2}\frac{1+u}{(1-u)}, && \text{(E.66)}
\end{aligned}$$

therefore, we have

$$U = \frac{(-1)^{2l+1}}{(1-u)^{(2l+2)}} \int_0^\infty \eta^{l+3/2} J_{l+1/2}\left(\frac{1}{2}\zeta\eta\right) \exp\left[-\eta\frac{1+u}{2(1-u)}\right] d\eta \tag{E.67}$$

$$= \frac{(-1)^{2l+1}}{(1-u)^{(2l+2)}} \frac{\Gamma(2l+3)}{\Gamma(l+3/2)} \left(\frac{\zeta}{4}\right)^{l+1/2} \left[\frac{2(1-u)}{1+u}\right]^{2l+3} \tag{E.68}$$

$$\times F\left(\frac{2l+3}{2}, \frac{2l+4}{2}; l+\frac{3}{2}; -\frac{1}{4}\zeta^2 4\left(\frac{1-u}{1+u}\right)^2\right)$$

$$= (-1)^{2l+1} \frac{4(2l+2)!}{\Gamma(l+3/2)} \zeta^{l+1/2} \frac{(1-u)}{(1+u)^{2l+3}} \tag{E.69}$$

$$\times F\left(l+\frac{3}{2}, l+2; l+\frac{3}{2}; -\zeta^2\left(\frac{1-u}{1+u}\right)^2\right).$$

We note that the hypergrometric series F is a degenerate one:

$$F\left(l+\frac{3}{2}, l+2; l+\frac{3}{2}; -\zeta^2\left(\frac{1-u}{1+u}\right)^2\right) = \left\{1+\zeta^2\left(\frac{1-u}{1+u}\right)^2\right\}^{-l-2} \tag{E.70}$$

$$= \left\{\frac{(1+u)^2+\zeta^2(1-u)^2}{(1+u)^2}\right\}^{-l-2} \tag{E.71}$$

$$= \left\{\frac{(1+u)^2}{(1+u)^2+\zeta^2(1-u)^2}\right\}^{l+2}, \tag{E.72}$$

and hence, Eq. E.69 becomes

$$U = (-1)^{2l+1} \frac{4(2l+2)!}{\Gamma(l+3/2)} \zeta^{l+1/2} \frac{(1-u)}{(1+u)^{2l+3}} \left\{\frac{(1+u)^2}{(1+u)^2+\zeta^2(1-u)^2}\right\}^{l+2} \tag{E.73}$$

$$= (-1)^{2l+1} \frac{4(2l+2)!}{\Gamma(l+3/2)} \zeta^{l+1/2} \frac{1-u^2}{(1+\zeta^2)^{l+2}\left[1+2\frac{(1-\zeta^2)}{(1+\zeta^2)}u+u^2\right]^{l+2}} \tag{E.74}$$

$$= A\frac{1-u^2}{[1+2xu+u^2]^{l+2}}, \tag{E.75}$$

where

$$x = \frac{1-\zeta^2}{1+\zeta^2}, \tag{E.76}$$

$$A = (-1)^{2l+1} \frac{4(2l+2)!}{\Gamma(l+3/2)} \frac{\zeta^{l+1/2}}{(1+\zeta^2)^{l+2}}. \tag{E.77}$$

This is the final step where we introduce another identity first, we recall the Gegenbauer polynomail identity as shown in Eq. E.31

$$Q_\nu \equiv \frac{1}{\left(1+ut+u^2\right)^\nu} \tag{E.78}$$

$$\equiv \sum_{k=0}^{\infty} C_k^\nu(t)\,u^k \tag{E.79}$$

By performing operation both side with $u^{-\nu+1}\frac{\partial}{\partial u}u^\nu$, we have

$$u^{-\nu+1}\frac{\partial}{\partial u}\frac{u^\nu}{\left(1+ut+u^2\right)^\nu} = u^{-\nu+1}\frac{\partial}{\partial u}u^\nu \sum_{k=0}^{\infty} C_k^\nu(t)\,u^k \tag{E.80}$$

$$u^{-\nu+1}\nu\frac{u^{\nu-1}+tu^\nu+u^{\nu+1}-tu^\nu-2u^{\nu+1}}{\left(1+ut+u^2\right)^{\nu+1}} = \sum_{k=0}^{\infty}(\nu+k)\,C_k^\nu(t)\,u^k \tag{E.81}$$

$$u^{-\nu+1}\nu\frac{u^{\nu-1}\left(1-u^2\right)}{\left(1+ut+u^2\right)^{\nu+1}} = \sum_{k=0}^{\infty}(\nu+k)\,C_k^\nu(t)\,u^k \tag{E.82}$$

$$\frac{\nu\left(1-u^2\right)}{\left(1+ut+u^2\right)^{\nu+1}} = \sum_{k=0}^{\infty}(\nu+k)\,C_k^\nu(t)\,u^k \tag{E.83}$$

Hence, we declare this as a new identity

$$\frac{\nu\left(1-u^2\right)}{\left(1+ut+u^2\right)^{\nu+1}} = \sum_{k=0}^{\infty}(\nu+k)\,C_k^\nu(t)\,u^k \tag{E.84}$$

By setting $\nu = l+1$ and $t = x$, we may rewrite our U function as

$$U = A\frac{1-u^2}{\left[1+2xu+u^2\right]^{l+2}} \tag{E.85}$$

$$= \frac{A}{l+1}\sum_{k=0}^{\infty}(l+1+k)\,C_k^{l+1}(x)\,u^k \tag{E.86}$$

$$= \frac{A}{l+1}\sum_{n=l+1}^{\infty} nC_{n-l-1}^{l+1}(x)\,u^{n-l-1}. \tag{E.87}$$

As a result, we finalize the definition of our U function

$$U = \sum_{n=l+1}^{\infty}\frac{I_{nl}(\zeta)}{(n+l)!}u^{n-l-1} = \frac{A}{l+1}\sum_{n=l+1}^{\infty} nC_{n-l-1}^{l+1}\left(\frac{1-\zeta^2}{1+\zeta^2}\right)u^{n-l-1}, \tag{E.88}$$

with

$$I_{nl}(\zeta) = \frac{An\,(n+l)!}{(l+1)}C_{n-l-1}^{l+1}\left(\frac{1-\zeta^2}{1+\zeta^2}\right).$$

Finally, the radial integral is solved, and the momentum transformation wavefunction is completed

$$\begin{aligned}
\Psi_{n,l,m}(P,\Theta,\Phi) &= A(2Y)^{-(l+5/2)} I_{nl}(\zeta) && \text{(E.89)}\\
&= A(2Y)^{-(l+5/2)} \frac{An(n+l)!}{(l+1)} C_{n-l-1}^{l+1}\left(\frac{1-\zeta^2}{1+\zeta^2}\right) && \text{(E.90)}\\
&= \left\{\frac{1}{(2\pi)^{1/2}} e^{\pm im\Phi}\right\}\left\{\left(\frac{(2l+1)(l-m)!}{2(l+m)!}\right)^{\frac{1}{2}} P_l^m(\cos\Theta)\right\} && \text{(E.91)}\\
&\quad\times\left\{\frac{B\zeta^l}{(1+\zeta^2)^{l+2}} C_{n-l-1}^{l+1}\left(\frac{1-\zeta^2}{1+\zeta^2}\right)\right\}
\end{aligned}$$

where $\zeta=\frac{P}{Y\hbar}, B=-(-1)^{2l+1}(-i)^l\,2^{2l+4}l!\frac{\pi}{Y^{3/2}}\left(\frac{n(n-l-1)!}{(n+l)!}\right)^{\frac{1}{2}}$ and $\Gamma(m+1/2)=\frac{(2m)!}{4^m m!}\sqrt{\pi}$.

Obviously, we can see that he imaginary part and negative factor can be omitted, and yielding the final expression of the momentum-space distribution of the hydrogen wavefunction.

$$\begin{aligned}
\Psi_{n,l,m}(P,\Theta,\Phi) &= \left\{\frac{1}{(2\pi)^{1/2}} e^{\pm im\Phi}\right\}\left\{\left(\frac{(2l+1)(l-m)!}{2(l+m)!}\right)^{\frac{1}{2}} P_l^m(\cos\Theta)\right\} && \text{(E.92)}\\
&\quad\times\left\{\frac{\pi 2^{2l+4} l!}{Y^{3/2}}\left(\frac{n(n-l-1)!}{(n+l)!}\right)^{\frac{1}{2}} \frac{\zeta^l}{(1+\zeta^2)^{l+2}} C_{n-l-1}^{l+1}\left(\frac{1-\zeta^2}{1+\zeta^2}\right)\right\}
\end{aligned}$$

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