

STUDY OF LASER INTERACTIONS WITH QUANTUM
PARTICLES

TAN KAI SHUEN

FACULTY OF SCIENCE
UNIVERSITY OF MALAYA
KUALA LUMPUR

2013

STUDY OF LASER INTERACTIONS WITH QUANTUM
PARTICLES

TAN KAI SHUEN

DISSERTATION SUBMITTED IN FULFILMENT
OF THE REQUIREMENTS
FOR THE DEGREE OF MASTER OF SCIENCE

DEPARTMENT OF PHYSICS
FACULTY OF SCIENCE
UNIVERSITY OF MALAYA
KUALA LUMPUR

2013

UNIVERSITI MALAYA

ORIGINAL LITERARY WORK DECLARATION

Name of Candidate: Tan Kai Shuen (I.C./Passport No.: 881011-56-6097)

Registration/Matrix No.: SGR110080

Name of Degree: Master of Science

Title of Project Paper/Research Report/Dissertation/Thesis ("this Work"): Study of Interactions Between Quantum Particles and Nanoparticle

Field of Study: Quantum and Laser Science

I do solemnly and sincerely declare that:

- (1) I am the sole author/writer of this Work;
- (2) This work is original;
- (3) Any use of any work in which copyright exists was done by way of fair dealing and for permitted purposes and any excerpt or extract from, or reference to or reproduction of any copyright work has been disclosed expressly and sufficiently and the title of the Work and its authorship have been acknowledged in this Work;
- (4) I do not have any actual knowledge nor do I ought reasonably to know that the making of this work constitutes an infringement of any copyright work;
- (5) I hereby assign all and every rights in the copyright to this Work to the University of Malaya ("UM"), who henceforth shall be owner of the copyright in this Work and that any reproduction or use in any form or by any means whatsoever is prohibited without the written consent of UM having been first had and obtained;
- (6) I am fully aware that if in the course of making this Work I have infringed any copyright whether intentionally or otherwise, I may be subject to legal action or any other action as may be determined by UM.

Candidate's Signature

Date

Subscribed and solemnly declared before,

Witness's Signature

Date

Name:

Designation:

ABSTRACT

In this research, we consider the interaction between an atomic or molecular quantum system (QS) and a metallic nanoparticle (MP). We modelled QS by a three-level lambda system. Under the probe field and control field, the QS become transparent to incident light within a narrow spectrum of frequency, a phenomena known as electromagnetically induced transparency (EIT). The quantum coherence of the QS under EIT and the strong exciton-plasmon coupling between the QS and MP are dependant on each other. The analytical expressions for the interaction of QS and MP are derived using two methods. First, the QS-MP system is considered as a one-dimensional problem. The expressions derived using this method is not general, however, they provide a simple and more intuitive understanding of the interaction. The second method employed to derive the analytical expressions is by using a vectorial description. Using this model, the exact and general expressions describing the QS-MP system are derived. The density matrix of the QS-MP system was used to obtain numerical results for the dielectric function of QS and MP. The effect of field detuning, distance between the QS and MP, laser field direction and polarization on the dielectric functions are investigated.

ABSTRAK

Penyelidikan ini mengkaji interaksi sistem kuantum atomik atau molekular (QS) dengan zarah nano (MP). Dalam penyelidikan ini QS dimodelkan oleh system lambda dengan tiga aras tenaga. Dengan menggunakan dua laser secara berasingan pada QS, QS akan menjadi lutsinar dalam dalam satu spektrum frekuensi yang sempit. Fenomena ini dikenali sebagai "electromagnetically induced transparency" (EIT). Koheren kuantum QS dengan kesan EIT dipengaruhi oleh gandingan exciton-plasmon QS dan MP. Persamaan analitikal untuk interaksi QS dan MP diperolehi melalui dua kaedah. Kaedah pertama mengkaji sistem QS-MP dalam satu dimensi. Persamaan-persamaan yang diperolehi melalui kaedah ini adalah tidak umum tetapi persamaan-persamaan ini dapat memberikan gambaran yang mudah dan senang difahami mengenai interaksi QS-MP. Dalam kaedah kedua, sistem QS-MP dikaji dalam model vektor. Kaedah ini memberikan persamaan yang tepat untuk sistem yang dikaji. Keputusan numerikal diperolehi untuk fungsi dielektrik QS melalui "density matrix" QS-MP. Kesan parameter seperti arah laser serta jarak QS dan MP terhadap fungsi dielektrik dikaji dalam penyelidikan ini.

ACKNOWLEDGEMENTS

I would like to thank my research advisor, associate professor Dr. Raymond Ooi for giving me the opportunity to perform research in his group. While working in the group, I have the chance to work on exciting research topics that are at the frontier of the fields of Physics. Regular discussion with Dr. Raymond helped me to keep tab of my research progress and help to ensure that I stay on the right track in my research work. With his help and guidance, I was motivated to work hard. I was able to generate and derive interesting and novel results, learn new physics and subsequently publishing a paper in a top tier journal.

I would like to thank my colleagues, Nor Hazmin and Ng Kam Seng. Through many helpful discussion with Nor, I was able to understand some of the Physics concepts better and thus helping to overcome some hurdles I faced during my research. Kam Seng on the other hand had helped me on many of the mathematical and programming problems I faced.

I would like to thank all the other members of the research group who helped in creating a fun and intellectual working environment. Especially, I would like to thank Ng Kam Seng and Seow Poh Choo for being great friends and giving support to me during my research period.

Last but not least, I thank my parents and my sister for their continual support in my study and research work.

TABLE OF CONTENTS

| | |
|--|-------------|
| ORIGINAL LITERARY WORK DECLARATION | ii |
| ABSTRACT | iii |
| ABSTRAK | iv |
| ACKNOWLEDGEMENTS | v |
| TABLE OF CONTENTS | vi |
| LIST OF FIGURES | vii |
| LIST OF APPENDICES | viii |
| CHAPTER 1: INTRODUCTION | 1 |
| 1.1 Literature Review | 1 |
| 1.1.1 Review of Quantum Coherence and Electromagnetically Induced Transparency | 1 |
| 1.1.2 Review of Hybrid System of Plasmonic Nanoparticle and Quantum System | 2 |
| 1.2 Motivation | 2 |
| CHAPTER 2: THEORETICAL BACKGROUND | 4 |
| 2.1 Density Matrix Formalism | 4 |
| 2.2 Two level atom | 7 |
| 2.2.1 Rotating Wave Approximation | 11 |
| 2.2.2 Two Level Atom in Density Matrix Formalism | 13 |
| 2.3 Electromagnetically-induced Transparency | 15 |
| CHAPTER 3: LINEAR MODEL FOR LOCAL FIELDS | 21 |
| 3.1 Local Field Equations For Quantum System and Metallic Nanoparticle | 21 |
| 3.1.1 Local Field in 1 Dimension | 24 |
| CHAPTER 4: VECTORIAL LOCAL FIELD | 28 |
| 4.1 Vectorial Local Field Equations For QS and MP | 28 |
| 4.1.1 Vectorial Local Fields in 2-dimension | 31 |
| CHAPTER 5: RESULTS AND DISCUSSION | 38 |
| CHAPTER 6: CONCLUSION | 43 |
| APPENDICES | 45 |
| REFERENCES | 56 |

LIST OF FIGURES

| | | |
|------------|---|----|
| Figure 2.1 | Electromagnetically induced transparency in three-level atom | 17 |
| Figure 3.1 | Configuration of a quantum system (QS) and a MP probed by a laser with field \mathbf{E} . The internal states of the QS described by a three-level system with $a - c$ transition driven by a strong external control laser. | 21 |
| Figure 5.1 | (The spectra for the real and imaginary parts of the dielectric function ϵ_{QS} versus R the spacing between the QS and MP for : a) $\epsilon_m = 0.453 + 3.35i$ with a small enhancement of $\gamma \simeq 3$. b) $\epsilon_m = -4.9 + 0.05i$ with $\gamma = 3.34 + 0.06i$ for Ag using $\omega_p = 9.1\text{eV} = 2.2 \times 10^{15}\text{s}^{-1}$, $\Gamma_m = 18\text{meV} = 2.73 \times 10^{13}\text{s}^{-1}$, $\epsilon_\infty = 3.7$ corresponding to $\omega_{SP} = 5.34 \times 10^{15}\text{s}^{-1}$, $\omega_{ab} = 2\pi c/(400\text{nm}) = 4.71 \times 10^{15}\text{s}^{-1}$. Other parameters are: $\epsilon_b = 1.5$, $\Omega_c = 5\Gamma$, the probe field amplitude $E_0 = 0.1\hbar\Gamma/\wp$, $\wp = 2 \times 10^{-29}\text{Cm}^{-1}$, where $\Gamma = 10^9\text{s}^{-1}$ (Folk, Marcus, & Harris, 2001) for the decay rate. | 38 |
| Figure 5.2 | (The spectra of ϵ_{QS} versus R for different ϕ, θ with surface plasmon resonance (SPR) condition with $\omega_{ab} = \omega_{SP} = 5.34 \times 10^{15}\text{s}^{-1}$, which gives a large enhancement, $\gamma = 0.88 + 131i$ at $\omega = \omega_{SP}$. All other parameters are the same as in Fig. 5.1b. | 40 |
| Figure 5.3 | (The spectra of $\text{Re } \epsilon_{QS}$ and $\text{Im } \epsilon_{QS}$ versus θ the incident angle of the probe laser with SPR for various phase angles $\phi = 0, \pi/4, \pi/2, 3\pi/4$ at $R = 15a$. Other parameters are the same as in Fig. 5.2. | 41 |

LIST OF APPENDICES

| | | |
|------------|--|----|
| Appendix A | Multipole Expansion | 46 |
| Appendix B | Deriving The Equations of Motion for The Wave Function | 52 |

CHAPTER 1

INTRODUCTION

1.1 Literature Review

1.1.1 Review of Quantum Coherence and Electromagnetically Induced Transparency

Since the advent of quantum mechanics in the early twentieth century, the concept of quantum coherence has been an active area of study and research. Laser provides means of preparing and manipulating quantum coherence in atomic and molecular media. These media exhibit quantum coherence through the interference of the different excitation paths of the system. The field of quantum coherence started with Hanle effect (Hanle, 1924; Alnis et al., 2003). Another early work of quantum coherence is coherent population trapping (CPT) (Whitley & Stroud, 1976). One way to achieve CPT is through quantum interference in a three-level system in which the system is in a coherent superposition of two states, both of which are coupled by lasers to a third state. This superposition state is called a dark state. Atoms that are pumped to the dark state are "trapped" in the state as the probability of absorbing a photon at this state is 0. Thus, the dark state does not interact with light and no fluorescence is observed.

The concept of CPT is closely related to the phenomenon of electromagnetically induced transparency (EIT). In EIT, two states in a three-level system is coupled to a third state through a weak probe field and a strong control field. Since in CPT, no photon will be absorbed in the dark state, thus there exists a narrow spectral region where the system is transparent. This effect is known as EIT. In the EIT regime, the optical properties of the medium is highly modified and lead to many interesting and counterintuitive phenomena. Many theoretical and experimental work has been devoted to the study of EIT. For example, professor Lene Hau demonstrated experimentally that through EIT in an ultracold gas of sodium atoms, optical pulses could be slowed to 17 metres per second (cycling speed) (Hau, Harris, Dutton, & Behroozi, 1999). On the other hand, professor Fleischauer and Lukin have identified theoretically the coupled excitations in EIT known as "dark-state

polaritons" and that the mixing angle between light and matter can be controlled by an external field to manipulate properties of the medium such as the group velocity of the propagating pulse (Fleischhauer & Lukin, 2000).

1.1.2 Review of Hybrid System of Plasmonic Nanoparticle and Quantum System

Recent work on studying the interactions of plasmonic effect on the quantum coherence and interference of quantum dots have shown interesting results. Here we highlight several important results and progress made in this field of research.

In a strongly coupled plasmon-quantum dot system, gain with inversion can be achieved through a change in plasma frequency (Hatef & Singh, 2010). The dissipation of metallic nanoparticle can be controlled through infrared laser in a metallic nanoparticle-semiconductor quantum dot system (Sadeghi, Deng, Li, & Huang, 2009). QED effects like vacuum Rabi-splitting was found by placing a semiconductor quantum dot in between two metallic nanoparticles (Savasta et al., 2010). The presence of a metallic film can also dramatically affects the fluorescence behavior of a nanocrystal quantum dot (Shimizu, Woo, Fisher, Eisler, & Bawendi, 2002).

1.2 Motivation

The study of the hybrid system of nanoparticle and quantum-dot has provided many interesting results and open up many possibilities. In this research, we would like to focus on yet another aspect of the hybrid system and try to gain new physics and insights to the hybrid system.

In this research we study the interaction between a quantum system (QS) and metallic-nanoparticle (MP). In particular, we are interested in the interdependence of the quantum coherence under electromagnetically-induced transparency of a quantum system (QS) modelled by a three-level lambda system and the strong exciton-plasmon coupling between QS and MP. Due to the long-range Coulomb interaction between QS and MP, there is a dipole-dipole interaction that couple the two systems and allow for excitation transfer between the systems.

Using a 1-dimensional linear model, we study the QS-MP system and gain insights on the near field behavior of the QS. For a more general description, we employ a vectorial model with arbitrary laser direction and look at how the optical properties of the QS is affected by the presence of MP. Finally we plot the results and study how the dispersion and absorption spectra of the QS is affected by different parameters such as laser direction, polarization and the interparticle distance.

CHAPTER 2

THEORETICAL BACKGROUND

2.1 Density Matrix Formalism

As we'll be employing density matrix formalism to describe our QS system and also eventually using it to solve for the equations of motion for the density matrix, it is useful to introduce briefly the density matrix concept.

The density operator for mixed state is defined as:

$$\hat{\rho}(t) = \sum_i p_i |\psi_i(t)\rangle \langle \psi_i(t)| \quad (2.1)$$

where p_i is the probability of that the system is in the state $|\psi_i(t)\rangle$. The density matrix is hermitian since p_i is real and $(|\psi_i(t)\rangle \langle \psi_i(t)|)^\dagger = |\psi_i(t)\rangle \langle \psi_i(t)|$. In the special case that all p_i vanish except for $i = j$, i.e $p_i = \delta_{ij}$, then $p_j = 1$ and the density matrix becomes:

$$\hat{\rho} = |\psi_j(t)\rangle \langle \psi_j(t)| \quad (2.2)$$

This is the density operator for a pure state.

Note that each $|\psi_i\rangle$ can be expanded in an orthonormal basis, e.g energy eigenstates. Thus, expanding $|\psi_i\rangle$ in energy basis $|n\rangle$:

$$\begin{aligned} |\psi_i(t)\rangle &= \sum_n |n\rangle \langle n | \psi_i(t)\rangle \\ &= \sum_n c_n^i |n\rangle \end{aligned} \quad (2.3)$$

Taking the expectation of the density matrix in the energy states:

$$\begin{aligned}
\langle n | \hat{\rho}(t) | m \rangle &= \sum_i \langle n | \Psi_i(t) \rangle p_i \langle \Psi_i(t) | m \rangle \\
&= \sum_i \sum_{q, q'} \langle n | c_q^i(t) | q \rangle p_i \langle q' | c_{q'}^{(i)*}(t) | m \rangle \\
&= \sum_i \sum_{q, q'} p_i c_q^i(t) c_{q'}^{(i)*}(t) \langle n | q \rangle \langle q' | m \rangle \\
&= \sum_i \sum_{q, q'} p_i c_q^i(t) c_{q'}^{(i)*}(t) \delta_{n, q} \delta_{q', m} \\
&= \sum_i p_i c_n^i(t) c_m^{(i)*}(t) \tag{2.4}
\end{aligned}$$

and in pure case:

$$\langle n | \hat{\rho}(t) | m \rangle = c_n(t) c_m^*(t) \tag{2.5}$$

Taking the trace of the matrix operator:

$$\begin{aligned}
Tr(\hat{\rho}) &= \sum_n \langle n | \hat{\rho} | n \rangle \\
&= \sum_n \sum_i p_i \langle n | \Psi_i(t) \rangle \langle \Psi_i(t) | n \rangle \\
&= \sum_n \sum_i p_i c_n^i c_n^{(i)*} \\
&= \sum_i p_i \sum_n |c_n^i|^2 \\
&= \sum_i p_i \\
&= 1 \tag{2.6}
\end{aligned}$$

where each of the state vector is assumed to be normalized, i.e $\langle \Psi_i(t) | \Psi_i(t) \rangle = 1$, thus for each i , $\sum_n |c_n^i|^2 = 1$. Since p_i is the probability of the system to be in state $|\Psi_i(t)\rangle$, thus the sum of probability, $\sum_i p_i$ has to be equal to 1. Also, since $\hat{\rho}$ is Hermitian, the diagonal elements $\langle n | \hat{\rho} | n \rangle$ must be real. For a particular n , $0 \leq |c_n^i|^2 \leq 1$, thus, $0 \leq \langle n | \hat{\rho} | n \rangle \leq 1$.

Now consider $Tr(\hat{\rho}^2)$. For a pure state, $\hat{\rho}^2 = |\Psi(t)\rangle \langle \Psi(t) | |\Psi(t)\rangle \langle \Psi(t) | = |\Psi(t)\rangle \langle \Psi(t) | = \hat{\rho}$, thus $Tr(\hat{\rho}^2) = Tr(\hat{\rho}) = 1$. For mixed states:

$$\begin{aligned}
Tr(\hat{\rho}^2) &= \sum_n \langle n | \hat{\rho}^2 | n \rangle \\
&= \sum_n \sum_i \sum_j p_i p_j \langle n | \psi_i(t) \rangle \langle \psi_i(t) | \psi_j(t) \rangle \langle \psi_j(t) | n \rangle \\
&= \sum_i \sum_j p_i p_j \sum_n \langle \psi_j(t) | n \rangle \langle n | \psi_i(t) \rangle \langle \psi_i(t) | \psi_j(t) \rangle \\
&= \sum_i \sum_j p_i p_j \left| \langle \psi_i(t) | \psi_j(t) \rangle \right|^2 \\
&\leq \left[\sum_i p_i \right]^2 = 1
\end{aligned} \tag{2.7}$$

where the closure relation was used, i.e $\sum_n |n\rangle \langle n| = \mathbf{I}$. Also, $0 \leq \left| \langle \psi_i(t) | \psi_j(t) \rangle \right|^2 \leq 1$ where the equality $\left| \langle \psi_i(t) | \psi_j(t) \rangle \right|^2 = 1$ holds only if all the $|\psi_i(t)\rangle$ are collinear in Hilbert space, i.e all the $|\psi_i(t)\rangle$ are equivalent up to an overall phase factor. In this case, $\hat{\rho}$ is a sum of same state vectors differing only by an overall phase factor. The superposition of pure state vectors is another pure state vector. Thus, the following criteria is true for pure and mixed states:

$$Tr(\hat{\rho}^2) = 1 \text{ for pure states} \tag{2.8}$$

$$Tr(\hat{\rho}^2) < 1 \text{ for mixed states} \tag{2.9}$$

For a particular state $|\psi_i(t)\rangle$, the state is a pure state. The expectation value is given by:

$$\begin{aligned}
\langle \hat{O} \rangle_i &= \sum_{n,m} \langle n | c_n^{(i)*}(t) \hat{O} c_m^i(t) | m \rangle \\
&= \sum_{n,m} c_m^i c_n^{(i)*} O_{nm} \\
&= \sum_{n,m} \rho_{mn}^i O_{nm}
\end{aligned} \tag{2.10}$$

where eq. (2.32) was used to get eq. (2.10). For the statistical mixture of $|\psi_i(t)\rangle$, the ensemble expectation value is given by:

$$\begin{aligned}
\langle \hat{O} \rangle &= \sum_i \sum_{n,m} p_i \langle n | c_n^{(i)*}(t) \hat{O} c_m^i(t) | m \rangle \\
&= \sum_i \sum_{n,m} p_i c_m^i c_n^{(i)*} O_{nm} \\
&= \sum_i \sum_{n,m} p_i \rho_{mn}^i O_{nm}
\end{aligned} \tag{2.11}$$

which is just a weighted sum of eq. (2.10) where the weight of each $|\psi_i(t)\rangle$ is p_i . It can also be shown that $\langle \hat{O} \rangle$ is equals to $Tr(\hat{\rho}\hat{O})$:

$$\begin{aligned}
Tr(\hat{\rho}\hat{O}) &= \sum_n \langle n | \hat{\rho}\hat{O} | n \rangle \\
&= \sum_n \sum_i p_i \langle n | \psi_i(t) \rangle \langle \psi_i(t) | \hat{O} | n \rangle \\
&= \sum_i p_i \langle \psi_i(t) | \hat{O} \sum_n | n \rangle \langle n | \psi_i(t) \rangle \\
&= \sum_i p_i \langle \psi_i(t) | \hat{O} | \psi_i(t) \rangle \\
&= \sum_i \sum_{n,m} p_i \langle n | c_n^{(i)*} \hat{O} c_m^i | m \rangle \\
&= \sum_i \sum_{n,m} p_i c_n^{(i)*} c_m^i O_{nm} \\
&= \sum_i \sum_{n,m} p_i \rho_{mn}^i O_{nm} \\
&= \langle \hat{O} \rangle
\end{aligned} \tag{2.12}$$

Again, the closure relation $\sum_n |n\rangle \langle n| = \mathbf{I}$ is used in the third line. $Tr(\hat{\rho}\hat{O}) = \langle \hat{O} \rangle$ means that if the density matrix of the system is known, then the expectation value of any operator can be calculated by taking the trace of the product of the operator in matrix representation with the density matrix in any order.

2.2 Two level atom

In our research we will be modelling the QS as three-level atoms. Here, we give a brief introduction to two-level system atom. The three-level system atom can be generalized from the two-level atom system.

Consider a two level system atom. An example of such a system is a spin-1 atom with two energy level interacting with a z-polarized field. The ground state of the system has total angular momentum number $J = 0$, while the excited state has total angular momentum number $J = -2, 0, 2$. In general all 3 of these sublevels can contribute to the resonant transition of the atom, however, if a z-polarized field is used, then the system is effectively two level as only the transition between the ground state and the $m_J = 0$ sublevel of the excited state contributes. For atoms that has non-integer spin, in general there can be a few sub-levels that can contribute to a resonant transition, however, the atom can still be restricted to two level using optical pumping techniques. Upon optical pumping, the atom is said to be oriented to a particular sublevel, this orientation depends on the frequency and polarization of the pump laser. (Paul R. Berman, 2011)

In the dipole approximation (refer to Appendix A for a detailed description of dipole approximation), the interaction Hamiltonian is given by:

$$\hat{V}(\mathbf{R}, t) \approx -\hat{\boldsymbol{\mu}} \cdot \mathbf{E}(\mathbf{R}, t) = e\hat{\mathbf{r}} \cdot \mathbf{E}(\mathbf{R}, t) \quad (2.13)$$

where $\hat{\boldsymbol{\mu}}$ is the dipole moment operator, $-e$ is the charge of the electron and \mathbf{R} is the nuclear position. If atomic motion is neglected then $\mathbf{R} = \mathbf{0}$.

The applied electric field is assumed to take the form:

$$\mathbf{E}(t) = \hat{\mathbf{z}}|E_0(t)|\cos[\omega t - \phi(t)] = \frac{1}{2}\hat{\mathbf{z}}|E_0(t)|[e^{i\phi(t)}e^{-i\omega t} + e^{-i\phi(t)}e^{i\omega t}] \quad (2.14)$$

where $\frac{1}{2}E_0(t) = \frac{1}{2}|E_0(t)|e^{i\phi(t)}$ is the positive frequency component of the field. $E_0(t) = |E_0(t)|e^{i\phi(t)}$ is the complex amplitude of the field, ω is the carrier frequency of the field and $\phi(t)$ is the phase of the field. The time-varying field amplitude forms the pulse envelope while the time varying phase factor gives a time-varying frequency to the field (chirp). Using this form of electric field, the interaction Hamiltonian becomes:

$$\hat{V}(\mathbf{R}, t) = e\hat{\mathbf{z}}|E_0(t)|\cos[\omega t - \phi(t)] \quad (2.15)$$

Denoting the ground state and excited state by $|1\rangle$ and $|2\rangle$, the probability amplitude of these two states can be written as column vector:

$$\mathbf{c} = \begin{pmatrix} c_1 \\ c_2 \end{pmatrix} \quad (2.16)$$

and the interaction Hamiltonian can be written in its matrix elements:

$$\begin{aligned} V_{12} &= ez_{12} |E_0(t)| \cos[\omega t - \phi(t)] \\ V_{21} &= ez_{21} |E_0(t)| \cos[\omega t - \phi(t)] \\ V_{11} &= 0 \\ V_{22} &= 0 \end{aligned} \quad (2.17)$$

where $z_{12} = \langle 1|\hat{z}|2\rangle = \langle 2|\hat{z}|1\rangle^* = z_{21}^*$. The diagonal element vanishes because operator \hat{z} has odd parity thus $\langle 1|\hat{z}|1\rangle$ and $\langle 2|\hat{z}|2\rangle$ each has an overall odd parity.

In general, the matrix elements are complex, but for any single transition element it can be taken as real with an appropriate choice of the phase factor in the wavefunction. Thus, by choosing the matrix elements to be real, the z-component dipole moment can be defined as:

$$ez_{12} = -(\mu_z)_{12} = ez_{21}^* \quad (2.18)$$

For the free atom Hamiltonian, it can also be written in terms of matrix elements:

$$\hat{H}_0 = \frac{\hbar}{2} \begin{pmatrix} -\omega_0 & 0 \\ 0 & \omega_0 \end{pmatrix} \quad (2.19)$$

where ω_0 is the transition frequency between $|1\rangle$ and $|2\rangle$. Then, the total Hamiltonian of the system is:

$$\begin{aligned} \hat{H}(t) &= \hat{H}_0 + \hat{V}(t) = \frac{\hbar}{2} \begin{pmatrix} -\omega_0 & 0 \\ 0 & \omega_0 \end{pmatrix} \\ &+ \hbar \begin{pmatrix} 0 & |\Omega_0(t)| \cos[\omega t - \phi(t)] \\ |\Omega_0^*(t)| \cos[\omega t - \phi(t)] & 0 \end{pmatrix} \end{aligned} \quad (2.20)$$

where $\Omega_0(t) = |\Omega_0(t)| e^{i\phi(t)} = -(\mu_z)_{12} \frac{E_0(t)}{\hbar}$ is defined as the Rabi frequency and $\phi(t)$ is the phase factor of the Rabi frequency. From the equation, it can be seen that the Rabi frequency is a measure of the atom-field coupling strength in frequency units. The Rabi frequency is defined such that $E_0(t)$, z_{12} and z_{21} are positive quantities.

Substituting in eq. (2.20) into eq. (B.11) derived in the appendix B:

$$i\hbar\dot{\mathbf{c}}(t) = \frac{\hbar}{2} \begin{pmatrix} -\omega_0 & 2|\Omega_0(t)| \cos[\omega t - \phi(t)] \\ 2|\Omega_0^*(t)| \cos[\omega t - \phi(t)] & \omega_0 \end{pmatrix} \mathbf{c}(t) \quad (2.21)$$

where

$$\mathbf{c}(t) = \begin{pmatrix} c_1(t) \\ c_2(t) \end{pmatrix} \quad (2.22)$$

Recall the Pauli spin matrices:

$$\hat{\sigma}_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \hat{\sigma}_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad \hat{\sigma}_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (2.23)$$

Then the Hamiltonian eq. (2.20) can be written in terms of Pauli spin matrices:

$$\hat{\mathbf{H}}(t) = -\frac{\hbar\omega_0}{2} \sigma_z + \hbar |\Omega_0(t)| \cos[\omega t - \phi(t)] \sigma_x \quad (2.24)$$

The Rabi frequency can be compared to the electronic transitions frequencies to get a sense of the coupling strength of the field and atom. Electronic transitions are usually of the order of 10^{14} - 10^{16} Hz (microwave to x-ray range), while for a field created by a typical cw wave laser having power on the order of a few *mW*, the Rabi frequency is on the order of MHz to GHz (about 8 order of magnitude less than the electronic transitions). The Rabi frequency will only be on the same order of magnitude with the electronic transitions at very intense pulses with power more than 10^{17} W/cm².

Since $\omega_0 \gg |\Omega_0(t)|$ typically, the resonance approximation or better known as the rotating wave approximation (RWA) is usually a good approximation for an atom.

2.2.1 Rotating Wave Approximation

Rotating wave approximation (RWA) is an approximation that eliminates terms in the Hamiltonian that are rapidly oscillating. RWA is closely related to the two-level approximation, it is assumed that the field frequency is near resonance with the atomic transition. RWA also has the additional condition that the field has low intensity so that the coupling strength is small, i.e $\omega_0 \gg |\Omega_0(t)|$. Assume that the amplitude $|\Omega_0(t)|$ and phase $\phi(t)$ are slowly varying in time on a time scale in order of ω^{-1} or larger. $|\Omega_0(t)|$ is the term that modulates the pulse. Thus, a slowly varying $|\Omega_0(t)|$ means that the pulse is broad in the temporal domain (a small modulation of the plane wave). This means that the pulse is approximately a continuous wave (quasi-monochromatic). This makes sense since if the pulse has a large spread of frequencies, then there will be a significant amount of frequencies that are far off resonance making the RWA a poor approximation.

Furthermore, assume that:

$$\left| \frac{\omega_0 - \omega}{\omega_0 + \omega} \right| \ll 1 \quad (2.25)$$

$$\left| \frac{\Omega_0(t)}{\omega_0 + \omega} \right| \ll 1 \quad (2.26)$$

Eq. (2.25) assumes that the amplitude of the detuning of the field, $|\delta| = |\omega_0 - \omega|$ is much smaller than the transition frequency, while eq. (2.26) assumes that the Rabi frequency is much smaller than the transition frequency. Using trigonometric relation:

$$\cos(\theta) = \frac{e^{i\theta} + e^{-i\theta}}{2} \quad (2.27)$$

and noting that $\Omega_0(t) = |\Omega_0(t)| e^{i\phi(t)}$, then eq. (2.21) becomes:

$$\begin{aligned}
i\hbar\dot{\mathbf{c}}(t) &= \frac{\hbar}{2} \begin{pmatrix} -\omega_0 & \Omega_0(t)e^{-i\phi(t)}[e^{i(\omega t - \phi(t))} + c.c.] \\ \Omega_0^*(t)e^{-i\phi(t)}[e^{i(\omega t - \phi(t))} + c.c.] & \omega_0 \end{pmatrix} \mathbf{c}(t) \\
&= \frac{\hbar}{2} \begin{pmatrix} -\omega_0 & \Omega_0(t)e^{-2i\phi(t)}e^{i\omega t} + \Omega_0(t)e^{-i\omega t} \\ \Omega_0(t)e^{-2i\phi(t)}e^{i\omega t} + \Omega_0(t)e^{-i\omega t} & \omega_0 \end{pmatrix} \mathbf{c}(t) \\
&= \frac{\hbar}{2} \begin{pmatrix} -\omega_0 & \Omega_0^*(t)e^{i\omega t} + \Omega_0(t)e^{-i\omega t} \\ \Omega_0^*(t)e^{i\omega t} + \Omega_0(t)e^{-i\omega t} & \omega_0 \end{pmatrix} \mathbf{c}(t) \quad (2.28)
\end{aligned}$$

Using eq. (B.13), the equivalent of eq. (2.28) in the interaction picture is:

$$\begin{aligned}
i\hbar\dot{\mathbf{C}}_m(t) &= \frac{\hbar}{2} \begin{pmatrix} 0 & [\Omega_0^*(t)e^{i\omega t} + \Omega_0(t)e^{-i\omega t}]e^{i\omega_m t} \\ [\Omega_0^*(t)e^{i\omega t} + \Omega_0(t)e^{-i\omega t}]e^{i\omega_m t} & 0 \end{pmatrix} \mathbf{C}(t) \\
&= \frac{\hbar}{2} \begin{pmatrix} 0 & [\Omega_0^*(t)e^{i\omega t} + c.c.]e^{i(\omega_m - \omega)t} \\ [\Omega_0^*(t)e^{i\omega t} + c.c.]e^{i(\omega_m - \omega)t} & 0 \end{pmatrix} \mathbf{C}(t) \\
&= \frac{\hbar}{2} \begin{pmatrix} 0 & \Omega_0(t)e^{i(\omega_0 + \omega)t} + \Omega_0^*(t)e^{i\delta t} \\ \Omega_0^*(t)e^{i(\omega_0 + \omega)t} + \Omega_0(t)e^{i\delta t} & 0 \end{pmatrix} \mathbf{C}(t) \quad (2.29)
\end{aligned}$$

where $\omega_0 = \omega_m - \omega_n$ and $\delta = \omega_0 - \omega$. It is assumed that $\omega_2 - \omega_1 > 0$ such that in eq. (2.29), $\omega_{mn} = \omega_{21} > 0$ and $\omega_{mn} = \omega_{12} < 0$. Recall that in RWA, it was assumed that $|\omega_0 + \omega|$ is much larger than the detuning of the field $|\delta|$ and the Rabi frequency $|\Omega_0(t)|$. Physically this means that the exponential term with $\omega_0 + \omega$ is oscillating much faster than the detuning and Rabi frequency exponential terms. At a time scale much larger than $\frac{1}{\omega_0 + \omega}$, the $\omega_0 + \omega$ will average to zero and thus, contribute much less than the slowly varying detuning and Rabi frequency exponential terms. In RWA approximation, then eq. (2.29) becomes:

$$i\hbar\dot{\mathbf{C}}_m(t) = \frac{\hbar}{2} \begin{pmatrix} 0 & \Omega_0^*(t)e^{i\delta t} \\ \Omega_0(t)e^{i\delta t} & 0 \end{pmatrix} \mathbf{C}(t) \quad (2.30)$$

Another way to describe a two-level system is using density matrix formalism. Using density matrix formalism, consider now a two-level atom using density matrix with state

vector defined as:

$$|\Psi(t)\rangle = c_1(t)|1\rangle + c_2(t)|2\rangle \quad (2.31)$$

2.2.2 Two Level Atom in Density Matrix Formalism

Recall that:

$$\langle n|\hat{\rho}(t)|m\rangle = c_n(t)c_m^*(t) \quad (2.32)$$

Then the density matrix projected on this state vector has matrix elements:

$$\rho_{11} = c_1c_1^* \text{ (probability of being in the lower level)} \quad (2.33)$$

$$\rho_{12} = c_1c_2^* \text{ (coherence)} \quad (2.34)$$

$$\rho_{21} = c_2c_1^* = \rho_{12}^* \text{ (coherence)} \quad (2.35)$$

$$\rho_{22} = c_2c_2^* \text{ (probability of being in the upper level)} \quad (2.36)$$

The off-diagonal elements ρ_{12} and ρ_{21} are called coherences as they are related to the relative phase of state $|1\rangle$ and $|2\rangle$. To see why, write $c_1 = a_1e^{i\phi_1}$ and $c_2 = a_2e^{i\phi_2}$, then $c_1c_2^* = a_1a_2^*e^{i(\phi_1-\phi_2)}$ and $c_2c_1^* = a_2a_1^*e^{i(\phi_2-\phi_1)}$, where $\phi_1 - \phi_2$ and $\phi_2 - \phi_1$ are the relative phases of the two states.

In matrix notation, the density matrix is:

$$\hat{\rho} = \begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix} = \begin{pmatrix} |c_1|^2 & c_1c_2^* \\ c_2c_1^* & |c_2|^2 \end{pmatrix} \quad (2.37)$$

Notice that the density matrix is the outer product of the state amplitudes:

$$\hat{\rho} = \begin{pmatrix} c_1 \\ c_2 \end{pmatrix} \begin{pmatrix} c_1^* & c_2^* \end{pmatrix} \quad (2.38)$$

Consider now the time evolution of the density operator:

$$\dot{\hat{\rho}} = \sum_i p_i \left(\left| \dot{\psi}(t) \right\rangle \langle \psi_i(t)| + |\psi_i(t)\rangle \left\langle \dot{\psi}(t) \right| \right) \quad (2.39)$$

From Schrodinger's equation:

$$\begin{aligned} i\hbar \left| \dot{\psi}_i(t) \right\rangle &= \hat{H} |\psi_i(t)\rangle \\ \left| \dot{\psi}_i(t) \right\rangle &= -\frac{i}{\hbar} \hat{H} |\psi_i(t)\rangle \end{aligned} \quad (2.40)$$

Substituting in eq. (2.40) to eq. (2.39):

$$\begin{aligned} \dot{\hat{\rho}}(t) &= \sum_i p_i \left(-\frac{i}{\hbar} H |\psi_i(t)\rangle \langle \psi_i(t)| + |\psi_i(t)\rangle \frac{i}{\hbar} \langle \psi_i(t)| H \right) \\ &= -\frac{i}{\hbar} \left(H \sum_i p_i |\psi_i(t)\rangle \langle \psi_i(t)| - \sum_i p_i |\psi_i(t)\rangle \langle \psi_i(t)| H \right) \\ &= -\frac{i}{\hbar} (H\rho - \rho H) \\ &= -\frac{i}{\hbar} [H, \rho] \end{aligned} \quad (2.41)$$

This equation is known as the von Neumann equation. The Hamiltonian of a two-level atom interacting with a field is given by:

$$\begin{aligned} \hat{H}(t) &= \hat{H}_0 + \hat{V}(t) \\ &= \frac{\hbar}{2} \begin{pmatrix} -\omega_0 & 0 \\ 0 & \omega_0 \end{pmatrix} \\ &\quad + \hbar \begin{pmatrix} 0 & |\Omega_0(t)| \cos[\omega t - \phi(t)] \\ |\Omega_0(t)| \cos[\omega t - \phi(t)] & 0 \end{pmatrix} \\ &= \frac{\hbar}{2} \begin{pmatrix} -\omega_0 & 2|\Omega_0(t)| \cos[\omega t - \phi(t)] \\ 2|\Omega_0(t)| \cos[\omega t - \phi(t)] & \omega_0 \end{pmatrix} \end{aligned} \quad (2.42)$$

In RWA, the Hamiltonian becomes:

$$\hat{H}(t) = \frac{\hbar}{2} \begin{pmatrix} -\omega_0 & \Omega_0^*(t)e^{i\omega t} \\ \Omega_0(t)e^{-i\omega t} & \omega_0 \end{pmatrix} \quad (2.43)$$

Substituting eq. (2.43) into eq. (2.41):

$$\begin{pmatrix} \dot{\rho}_{11} & \dot{\rho}_{12} \\ \dot{\rho}_{21} & \dot{\rho}_{22} \end{pmatrix} = -\frac{i}{2} \begin{pmatrix} -\omega_0 & \Omega_0^*(t)e^{i\omega t} \\ \Omega_0(t)e^{-i\omega t} & \omega_0 \end{pmatrix} \begin{pmatrix} \rho_{11}(t) & \rho_{12}(t) \\ \rho_{21}(t) & \rho_{22}(t) \end{pmatrix} + \frac{i}{2} \begin{pmatrix} \rho_{11}(t) & \rho_{12}(t) \\ \rho_{21}(t) & \rho_{22}(t) \end{pmatrix} \begin{pmatrix} -\omega_0 & \Omega_0^*(t)e^{i\omega t} \\ \Omega_0(t)e^{-i\omega t} & \omega_0 \end{pmatrix} \quad (2.44)$$

Defining $\chi(t) = \frac{\Omega_0(t)}{2}$, then in terms of each of the element:

$$\begin{aligned} \dot{\rho}_{11}(t) &= -i\chi^*(t)e^{i\omega t}\rho_{21}(t) + i\chi(t)e^{-i\omega t}\rho_{12}(t) \\ \dot{\rho}_{22}(t) &= i\chi^*(t)e^{i\omega t}\rho_{21}(t) - i\chi(t)e^{-i\omega t}\rho_{12}(t) \\ \dot{\rho}_{12}(t) &= i\omega_0\rho_{12}(t) - i\chi^*(t)e^{i\omega t}[\rho_{22}(t) - \rho_{11}(t)] \\ \dot{\rho}_{21}(t) &= -i\omega_0\rho_{21}(t) + i\chi(t)e^{-i\omega t}[\rho_{22}(t) - \rho_{11}(t)] \end{aligned} \quad (2.45)$$

For a given $\chi(t)$, these equations can be solved. These equations can also be solved by finding the amplitudes and substituting it into the equations, e.g $\rho_{11} = |c_1|^2$, $\rho_{21} = c_2c_1^*$ etc.

2.3 Electromagnetically-induced Transparency

Finally, we are ready to look at the three-level system atom. Consider the semi-classical case of a three-level atom with state $|a\rangle$, $|b\rangle$ and $|c\rangle$ interacting with a single-mode field. The atom is treated quantum mechanically while the field is treated classically. From closure relation:

$$|a\rangle\langle a| + |b\rangle\langle b| + |c\rangle\langle c| = 1 \quad (2.46)$$

Multiplying eq. (2.46) to the unperturbed Hamiltonian:

$$\begin{aligned}
\hat{H}_0 &= \hat{H}_0(|a\rangle\langle a| + |b\rangle\langle b| + |c\rangle\langle c|) \\
&= \hbar\omega_a |a\rangle\langle a| + \hbar\omega_b |b\rangle\langle b| + \hbar\omega_c |c\rangle\langle c| \\
&= \sum_{n=a,b,c} \hbar\omega_n |n\rangle\langle n|
\end{aligned} \tag{2.47}$$

where $\hbar\omega_n$ ($n = a, b, c$) are the eigen-energies of the unperturbed Hamiltonian. Using electric dipole approximation, we assume that the wavelength of the electromagnetic radiation which induces, or is emitted during, transitions between different atomic energy levels is much larger than the typical size of a light atom. For a detailed description of the electric dipole approximation, refer to Appendix A. The interaction Hamiltonian written using dipole approximation is given by:

$$\hat{V}(\mathbf{R}, t) \approx -\hat{\boldsymbol{\mu}} \cdot \mathbf{E}(\mathbf{R}, t) = e\mathbf{r} \cdot \mathbf{E}(\mathbf{R}, t) \tag{2.48}$$

where $\hat{\boldsymbol{\mu}}$ is the dipole moment operator, $-e$ is the charge of the electron and \mathbf{R} is the nuclear position. If atomic motion is neglected then $\mathbf{R} = \mathbf{0}$. Again using closure relation, eq. (2.48) becomes

$$\begin{aligned}
\hat{V}(\mathbf{R}, t) &= e(|a\rangle\langle a| + |b\rangle\langle b| + |c\rangle\langle c|)r(|a\rangle\langle a| + |b\rangle\langle b| + |c\rangle\langle c|) \cdot \mathbf{E}(\mathbf{R}, t) \\
&= e[|a\rangle\langle a|r|b\rangle\langle b| + |a\rangle\langle a|r|c\rangle\langle c| + |b\rangle\langle b|r|a\rangle\langle a| + \\
&\quad + |b\rangle\langle b|r|c\rangle\langle c| + |c\rangle\langle c|r|a\rangle\langle a| + |c\rangle\langle c|r|b\rangle\langle b|] \cdot \mathbf{E}(\mathbf{R}, t) \\
&= -[\boldsymbol{\mu}_{ab}|a\rangle\langle b| + \boldsymbol{\mu}_{ac}|a\rangle\langle c| + \boldsymbol{\mu}_{bc}|b\rangle\langle c| + c.c.] \cdot \mathbf{E}(\mathbf{R}, t) \\
&= -[\boldsymbol{\mu}_{ab}|a\rangle\langle b| + \boldsymbol{\mu}_{ac}|a\rangle\langle c| + \boldsymbol{\mu}_{bc}|b\rangle\langle c| + c.c.] \cdot \mathbf{E}(\mathbf{R}, t) \\
&= -[\boldsymbol{\mu}_{ab}|a\rangle\langle b| + \boldsymbol{\mu}_{ac}|a\rangle\langle c| + c.c.] \cdot \mathbf{E}(\mathbf{R}, t)
\end{aligned} \tag{2.49a}$$

where $\langle i|r|j\rangle = r_{ij} = \langle j|r|i\rangle^* = r_{ji}^*$ ($i, j = a, b, c$) and $er_{ij} = -\boldsymbol{\mu}_{ij} = er_{ji}^*$; $\boldsymbol{\mu}_{ij}$ is the dipole moment between state i and j . The diagonal elements, r_{ii} vanishes since r has an odd parity and thus, the overall parity for r_{ii} is odd. Using selection rules, we find that the transition of $|b\rangle$ and $|c\rangle$ is dipole forbidden thus we set $\boldsymbol{\mu}_{bc} = 0$.

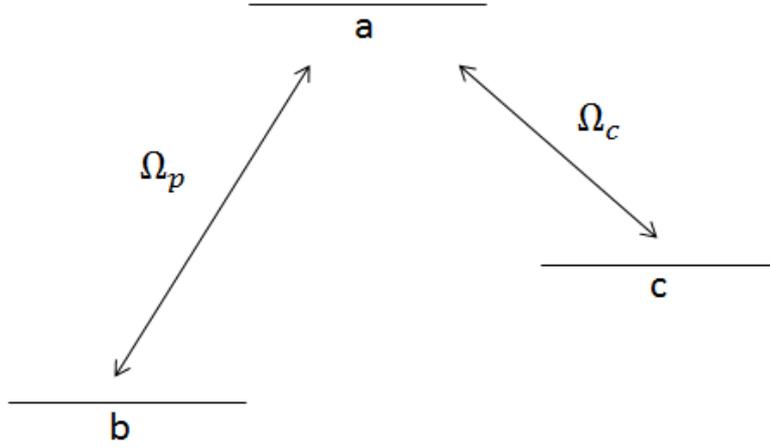


Figure 2.1: Electromagnetically induced transparency in three-level atom

Now consider having two fields, the probe field and control field (Ω_p, Ω_c) interacting with the three-level atom as shown in Fig. 2.1. We can tune the polarization of the field so that the probe field is coupled to $|b\rangle$ and $|a\rangle$ while the control field is coupled to $|a\rangle$ and $|c\rangle$. Then, eq. (2.49a) can be written separately for these 2 fields (Marlan O. Scully, 1997) :

$$\begin{aligned} \mathbf{V}(\mathbf{R}, t) &= -[\mu_{ab} |a\rangle \langle b| + \mu_{ac} |a\rangle \langle c| + adj] \cdot \mathbf{E}(\mathbf{R}, t) \\ &= -(\mu_{ab} |a\rangle \langle b| + adj) \cdot \mathbf{E}_p(\mathbf{R}, t) + (\mu_{ac} |a\rangle \langle c| + adj) \cdot \mathbf{E}_c(\mathbf{R}, t) \quad (2.50) \end{aligned}$$

with $\mathbf{E}(\mathbf{R}, t) = \mathbf{E}_p(\mathbf{R}, t) + \mathbf{E}_c(\mathbf{R}, t)$ and $\mathbf{E}_p(\mathbf{R}, t) = \mathbf{E}_{p0} e^{i(k_p z - \omega_p t)} + \mathbf{E}_{p0}^* e^{-i(k_p z - \omega_p t)}$, $\mathbf{E}_{c0}(\mathbf{R}, t) = \mathbf{E}_{c0} e^{i(k_c z - \omega_c t)} + \mathbf{E}_{c0}^* e^{-i(k_c z - \omega_c t)}$. Using the interaction picture representation we have:

$$\begin{aligned}
\widehat{\mathbf{V}}(\mathbf{R}, t) &= -[\mu_{ab}|a\rangle\langle b|e^{i\omega_{ab}t} + \mu_{ac}|a\rangle\langle c|] \cdot \mathbf{E}(\mathbf{R}, t) \\
&= -(\mu_{ab}|a\rangle\langle b|e^{i\omega_{ab}t} + adj) \cdot (\mathbf{E}_{p0}e^{i(k_p z - \omega_p t)} + \mathbf{E}_{p0}^*e^{-i(k_p z - \omega_p t)}) \\
&\quad - (\mu_{ac}|a\rangle\langle c|e^{i\omega_{ac}t} + adj) \cdot (\mathbf{E}_{c0}e^{i(k_c z - \omega_c t)} + \mathbf{E}_{c0}^*e^{-i(k_c z - \omega_c t)}) \\
&= -(\mu_{ab}|a\rangle\langle b|e^{i\omega_{ab}t} + adj)\mathbf{E}_{p0}e^{i(k_p z - \omega_p t)} \\
&\quad + (\mu_{ab}|a\rangle\langle b|e^{i\omega_{ab}t} + adj)\mathbf{E}_{p0}^*e^{-i(k_p z - \omega_p t)} \\
&\quad - (\mu_{ac}|a\rangle\langle c|e^{i\omega_{ac}t} + adj)\mathbf{E}_{c0}e^{i(k_c z - \omega_c t)} \\
&\quad + (\mu_{ac}|a\rangle\langle c|e^{i\omega_{ac}t} + adj)\mathbf{E}_{c0}^*e^{-i(k_c z - \omega_c t)} \\
&\simeq -(\mu_{ab}|a\rangle\langle b|e^{i\Delta_{ab}t} + adj)\mathbf{E}_{p0}e^{ik_p z} \\
&\quad - (\mu_{ac}|a\rangle\langle c|e^{i\Delta_{ac}t} + adj)\mathbf{E}_{c0}e^{ik_c z} \tag{2.51}
\end{aligned}$$

where $\Delta_{ab} = \omega_{ab} - \omega_p$ and $\Delta_{ac} = \omega_{ac} - \omega_c$. In the last step we have used RWA and eliminated the rapid oscillating terms.

Recall eq. (2.41), if we include the decay of the atomic state in to the quantum system, then the equation is modified:

$$\dot{\widehat{\rho}}(t) = -\frac{i}{\hbar} [\widehat{H}, \widehat{\rho}] - \Gamma \widehat{\rho} \tag{2.52}$$

where Γ is the decay operator. In terms of matrix elements, the decay operator can be written as:

$$\Gamma_{ij} = \frac{1}{2} (\gamma_i + \gamma_j) + \gamma_{ij} \tag{2.53}$$

where γ_i and γ_j are the population decay rate of state $|i\rangle$ and $|j\rangle$ while γ_{ij} is due to phase relaxation. γ_i , γ_j and γ_{ij} are related to the longitudinal (T_1) and transverse (T_2) relaxation time through:

$$\begin{aligned}
\gamma_i &= \frac{1}{T_1} \\
\gamma_{ij} &= \frac{1}{T_2} \tag{2.54}
\end{aligned}$$

and $\gamma_{ij} \neq 0$ only when $i \neq j$.

Define the three-level density matrix in slowly-varying amplitudes (Mikhailov, 2003):

$$\begin{aligned}
\tilde{\rho}_{aa}(t) &= \rho_{aa} \\
\tilde{\rho}_{bb}(t) &= \rho_{bb} \\
\tilde{\rho}_{cc}(t) &= \rho_{cc} \\
\tilde{\rho}_{ab}(t) &= \rho_{ab} e^{-i\omega_p t} \\
\tilde{\rho}_{ac}(t) &= \rho_{ac} e^{-i\omega_c t} \\
\tilde{\rho}_{bc}(t) &= \rho_{bc} e^{-i(\omega_p - \omega_c)t}
\end{aligned} \tag{2.55}$$

Under rotating wave approximation, the equations of motion for the density matrix elements are given by:

$$\begin{aligned}
\dot{\rho}_{aa} &= -i\Omega_p^* \rho_{ab} + i\Omega_p \rho_{ba} - i\Omega_d^* \rho_{ac} + i\Omega_c \rho_{ca} - 2\gamma \rho_{aa} \\
\dot{\rho}_{bb} &= i\Omega_p^* \rho_{ab} - i\Omega_p \rho_{ba} + \gamma \rho_{aa} - \gamma_{bc} \rho_{bb} + \gamma_{bc} \rho_{cc} \\
\dot{\rho}_{cc} &= i\Omega_p^* \rho_{ab} - i\Omega_p \rho_{ba} + \gamma \rho_{aa} - \gamma_{bc} \rho_{bb} + \gamma_{bc} \rho_{cc} \\
\dot{\rho}_{ab} &= -\Gamma_{ab} \rho_{ab} + i\Omega_p (\rho_{bb} - \rho_{aa}) + i\Omega_c \rho_{cb} \\
\dot{\rho}_{ca} &= -\Gamma_{ca} \rho_{ca} + i\Omega_c^* (\rho_{aa} - \rho_{cc}) - i\Omega_p^* \rho_{cb} \\
\dot{\rho}_{cb} &= -\Gamma_{cb} \rho_{cb} - i\Omega_p \rho_{ca} + i\Omega_c^* \rho_{ab}
\end{aligned} \tag{2.56}$$

where γ is the off-diagonal decay rate of ρ_{ab} and ρ_{ca} , γ_{bc} is the off-diagonal decay rates of ρ_{bc} and

$$\begin{aligned}
\Gamma_{ab} &= \Gamma_{ba}^* = \gamma + i\Delta_{bc} \\
\Gamma_{ca} &= \Gamma_{ac}^* = \gamma - i\Delta_{ab} \\
\Gamma_{cb} &= \Gamma_{bc}^* = \gamma_{bc} + i(\Delta_{bc} - \Delta_{ac})
\end{aligned} \tag{2.57}$$

In the steady state regime ($\dot{\rho}_{ij} = 0$), the solution to these equations are given by:

$$\rho_{ab} = i\Omega_p \frac{(\rho_{bb} - \rho_{aa}) (\Gamma_{ac}^* \Gamma_{cb} + |\Omega_p|^2) + |\Omega_c|^2 (\rho_{aa} - \rho_{cc})}{\Gamma_{ab} \Gamma_{ac}^* \Gamma_{cb} + \Gamma_{ac}^* |\Omega_c|^2 + \Gamma_{ab} |\Omega_p|^2} \quad (2.58)$$

$$\rho_{ca} = i\Omega_c^* \frac{(\rho_{aa} - \rho_{cc}) (\Gamma_{ab} \Gamma_{cb} + |\Omega_c|^2) + |\Omega_p|^2 (\rho_{bb} - \rho_{aa})}{\Gamma_{ab} \Gamma_{ac}^* \Gamma_{cb} + \Gamma_{ac}^* |\Omega_c|^2 + \Gamma_{ab} |\Omega_p|^2} \quad (2.59)$$

$$\rho_{cb} = i\Omega_p \Omega_c^* \frac{(\rho_{aa} - \rho_{cc}) \Gamma_{ab} - (\rho_{bb} - \rho_{aa}) \Gamma_{ac}^*}{\Gamma_{ab} \Gamma_{ac}^* \Gamma_{cb} + \Gamma_{ac}^* |\Omega_c|^2 + \Gamma_{ab} |\Omega_p|^2} \quad (2.60)$$

Using eq. (2.58)- eq. (2.60), we can find the steady state solution of density matrix for the system of interest.

CHAPTER 3

LINEAR MODEL FOR LOCAL FIELDS

3.1 Local Field Equations For Quantum System and Metallic Nanoparticle

For our research, we will be studying the interaction of quantum system (QS) and metallic nanoparticle (MP). In this chapter, we will derive the analytical expressions describing the local field of QS and MP in the near field limit. Then we will proceed to discuss how the local field depends on the various parameters. The expressions in near field limit will provide a simple and intuitive understanding of the interaction. In the next chapter, we will derive the exact and general expression for the local field of QS and MP.

The Hamiltonian of the QS is given by:

$$\hat{H} = \hat{H}_0 + \hbar (\Omega_b e^{-i\nu t} |a\rangle \langle b| + \Omega_c e^{-i\nu_c t} |a\rangle \langle c| + h.c) \quad (3.1)$$

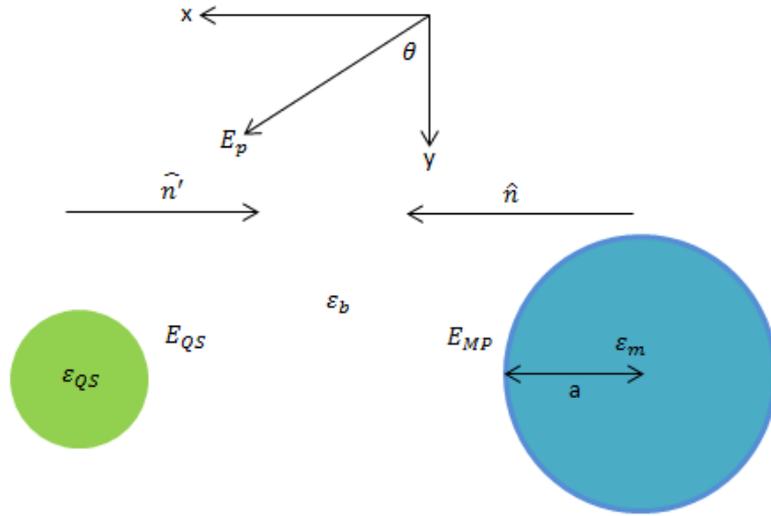


Figure 3.1: Configuration of a quantum system (QS) and a MP probed by a laser with field \mathbf{E} . The internal states of the QS described by a three-level system with $a - c$ transition driven by a strong external control laser.

The local electric field \mathbf{E}_{QS} is due to the probe field, control field and the polarization field from MP. The QS-MP scheme is shown in Fig 3.1. In the following we write separately the local field in terms of the contribution from probe field and control field.

$$\mathbf{E}_{QS,p} = \mathbf{E}_p e^{iv_p t} + \frac{1}{4\pi\epsilon_0 \epsilon_{effM,p} R^3} \mathbf{p}_{MP,p} \quad (3.2)$$

$$= E_0 e^{iv_p t} \hat{e} + \frac{1}{4\pi\epsilon_0 \epsilon_{effM,p} R^3} p_{MP,p} \hat{s}$$

$$\mathbf{E}_{QS,c} = \mathbf{E}_c e^{iv_c t} + \frac{1}{4\pi\epsilon_0 \epsilon_{effM,c} R^3} \mathbf{p}_{MP,c} \quad (3.3)$$

$$= E'_0 e^{iv_c t} \hat{e} + \frac{1}{4\pi\epsilon_0 \epsilon_{effM,c} R^3} p_{MP,c} \hat{s}$$

$$\epsilon_{effM,p} = \frac{2\epsilon_b + \epsilon_{MP,p}(\omega)}{3\epsilon_b} \quad (3.4)$$

$$\epsilon_{effM,c} = \frac{2\epsilon_b + \epsilon_{MP,c}(\omega)}{3\epsilon_b} \quad (3.5)$$

where the unit vector is $\hat{e} = (\sin \theta, e^{i\phi} \cos \theta)$; θ and ϕ determine the direction and the polarization of the probe field, respectively, \hat{s} is the direction of the field due to the polarization of MP and is given by $\hat{s} = 3\hat{n}(\hat{u}_m \cdot \hat{n}) - \hat{u}_m$ where \hat{u}_m is the unit vector of the MP dipole. ϵ_b and ϵ_{MP} are the background dielectric constant and the metallic nanoparticle dielectric constant respectively; ϵ_b depends on the system environment. Eq. (3.2) is the probe field contribution part for the local field; the first term on the RHS is the probe field and the 2nd term on the RHS is the polarization field of MP due to probe field. Similarly, eq. (3.3) is the control field contribution part for the local field; the first term on the RHS is the control field and the 2nd term on the RHS is the polarization field of MP due to control field.

The polarization of the MP is given by:

$$\mathbf{p}_{MP,p} = \alpha_{MP} \mathbf{E}_{MP,p} \quad (3.6)$$

$$\mathbf{p}_{MP,c} = \alpha_{MP} \mathbf{E}_{MP,c} \quad (3.7)$$

α_{MP} is the polarizability of the MP given by:

$$\alpha_{MP} = \frac{1}{K} \gamma a^3 \quad (3.8)$$

where γ is the plasmonic enhancement factor and we can find the expression for γ using Clausius-Mossotti relation:

$$\gamma = \frac{\epsilon_m(\omega) - \epsilon_b}{\epsilon_m(\omega) + 2\epsilon_b} \quad (3.9)$$

The linear response of the metal, ϵ_m may be described by:

$$\epsilon_m(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\Gamma)} \quad (3.10)$$

If we take the ensemble average of the polarization, then we find:

$$p_{QS,p} \simeq \mu(\rho_{ab} + \rho_{ba}) \quad (3.11)$$

$$p_{QS,c} \simeq \mu(\rho_{bc} + \rho_{cb}) \quad (3.12)$$

Similarly, the local field of the MP is also written in terms of the contribution from probe field and control field.

$$\begin{aligned} \mathbf{E}_{MP,p} &= \mathbf{E}_p + \frac{1}{4\pi\epsilon_0} \frac{\mathbf{p}_{QS,p}}{\epsilon_{effQ,p}R^3} \\ &= E_0 e^{iv_p t} \hat{e} + \frac{1}{4\pi\epsilon_0} \frac{p_{QS,p} \hat{s}'}{\epsilon_{effQ,p}R^3} \end{aligned} \quad (3.13)$$

$$\begin{aligned} \mathbf{E}_{MP,c} &= \mathbf{E}_c + \frac{1}{4\pi\epsilon_0} \frac{\mathbf{p}_{QS,c}}{\epsilon_{effQ,c}R^3} \\ &= E'_0 e^{iv_c t} \hat{e} + \frac{1}{4\pi\epsilon_0} \frac{p_{QS,c} \hat{s}'}{\epsilon_{effQ,c}R^3} \end{aligned} \quad (3.14)$$

$$\epsilon_{effQ,p} = \frac{2\epsilon_b + \epsilon_{QS,p}}{3\epsilon_b} \quad (3.15)$$

$$\epsilon_{effQ,c} = \frac{2\epsilon_b + \epsilon_{QS,c}}{3\epsilon_b} \quad (3.16)$$

where \hat{s}' is the direction of the field due to QS and is given by $\hat{s}' = 3\hat{n}(\hat{u}_q \cdot \hat{n}) - \hat{u}_q$. \hat{u}_q and \hat{u}_q is the unit vector of the dipole moment in QS.

In the specific case that the probe field is parallel with \hat{x} , then eq. (3.2) and eq. (3.13) can be rewritten as:

$$\begin{aligned}
\mathbf{E}_{QS,p} &= E_0 e^{i\nu_p t} \hat{e} + K \frac{p_{MP,p} \hat{s}}{\epsilon_{effM,p} R^3} \\
E_{QS,p\hat{x}} &= E_0 e^{i\nu_p t} \hat{x} + K \frac{p_{MP,p} (2\hat{x})}{\epsilon_{effM,p} R^3}
\end{aligned} \tag{3.17}$$

where

$$\begin{aligned}
\hat{s} &= 3\hat{n}(\hat{u}_m \cdot \hat{n}) - \hat{u}_m \\
&= 3(-\hat{x})(\hat{x} \cdot -\hat{x}) - \hat{x} \\
&= 2\hat{x}
\end{aligned} \tag{3.18}$$

$E_{QS,p}$ is in the \hat{x} direction since both the probe field and MP dipole field are in the \hat{x} direction.

$$\begin{aligned}
\mathbf{E}_{MPp} &= E_0 e^{i\nu_p t} \hat{e} + K \frac{p_{QS,p} \hat{s}'}{\epsilon_{effQ,p} R^3} \\
E_{MP,p\hat{x}} &= E_0 e^{i\nu_p t} \hat{x} + K \frac{p_{QS,p} (2\hat{x})}{\epsilon_{effQ,p} R^3}
\end{aligned} \tag{3.19}$$

where

$$\begin{aligned}
\hat{s}' &= 3\hat{n}'(\hat{u}_q \cdot \hat{n}') - \hat{u}_q \\
&= 3\hat{x}(\hat{x} \cdot \hat{x}) - \hat{x} \\
&= 2\hat{x}
\end{aligned} \tag{3.20}$$

3.1.1 Local Field in 1 Dimension

Recall that $\mathbf{p}_{MPp} = \alpha_{MP} \mathbf{E}_{MPp}$, $\alpha_{MP} = \frac{1}{K} \gamma a^3$ and $p_{QS,p} = \wp_{ba} \rho_{ab}$. Equating eq. (3.17) and eq. (3.19):

$$\begin{aligned}
E_{QS,p\hat{x}} - 2K \frac{P_{MP,p\hat{x}}}{\epsilon_{effM,p} R^3} &= E_{MP,p\hat{x}} - 2K \frac{P_{QS,p\hat{x}}}{\epsilon_{effQ,p} R^3} \\
E_{QS,p} &= E_{MP,p} - 2K \frac{P_{QS,p}}{\epsilon_{effQ,p} R^3} + 2K \frac{P_{MP,p}}{\epsilon_{effM,p} R^3} \\
&= E_{MP,p} - 2K \frac{\wp_{ba} \rho_{ab}}{\epsilon_{effQ,p} R^3} + 2 \frac{\gamma a^3}{\epsilon_{effM,p} R^3} E_{MP,p} \\
&= \left(1 + 2 \frac{\gamma a^3}{\epsilon_{effM,p} R^3}\right) E_{MP,p} - 2K \frac{\wp_{ba} \rho_{ab}}{\epsilon_{effQ,p} R^3} \\
&= \left(1 + 2 \frac{\gamma a^3}{\epsilon_{effM,p} R^3}\right) \left(E_0 e^{i\nu_p t} + 2K \frac{P_{QS,p}}{\epsilon_{effQ,p} R^3}\right) \\
&\quad - 2K \frac{\wp_{ba} \rho_{ab}}{\epsilon_{effQ,p} R^3} \\
&= \left(1 + 2 \frac{\gamma a^3}{\epsilon_{effM,p} R^3}\right) E_0 e^{i\nu_p t} + 2K \frac{\wp_{ba} \rho_{ab}}{\epsilon_{effQ,p} R^3} \\
&\quad + 4K \frac{\wp_{ba} \rho_{ab} \gamma a^3 \hat{x}}{\epsilon_{effM,p} \epsilon_{effQ,p} R^6} - 2K \frac{\wp_{ba} \rho_{ab}}{\epsilon_{effQ,p} R^3} \\
&= \left(1 + 2 \frac{\gamma a^3}{\epsilon_{effM,p} R^3}\right) E_0 e^{i\nu_p t} + 4K \frac{\wp_{ba} \rho_{ab} \gamma a^3 \hat{x}}{\epsilon_{effM,p} \epsilon_{effQ,p} R^6} \\
&= E_{p,eff} + 4K \frac{\wp_{ba} \rho_{ab} \gamma a^3 \hat{x}}{\epsilon_{effM,p} \epsilon_{effQ,p} R^6} \tag{3.21}
\end{aligned}$$

where $E_{p,eff} = \left(1 + 2 \frac{\gamma a^3}{\epsilon_{effM,p} R^3}\right) E_0 e^{i\nu_p t}$ is the effective probe field. Similarly, we can rewrite and combine the control field part:

$$\begin{aligned}
E_{QS,c\hat{x}} - 2K \frac{P_{MP,c\hat{x}}}{\epsilon_{effM,c} R^3} &= 2K \frac{P_{MP,c\hat{x}}}{\epsilon_{effM,c} R^3} = E_{MP,c\hat{x}} - 2K \frac{P_{QS,c\hat{x}}}{\epsilon_{effQ,c} R^3} \\
&= E_{MP,c} - 2K \frac{\wp_{ca} \rho_{ac}}{\epsilon_{effQ,c} R^3} + 2 \frac{\gamma a^3}{\epsilon_{effM,c} R^3} E_{MP,c} \\
&= \left(1 + 2 \frac{\gamma a^3}{\epsilon_{effM,c} R^3}\right) \left(E'_0 e^{i\nu_c t} + 2K \frac{\wp_{ca} \rho_{ac}}{\epsilon_{effQ,c} R^3}\right) \\
&\quad - 2K \frac{\wp_{ca} \rho_{ac}}{\epsilon_{effQ,c} R^3} \\
&= \left(1 + 2 \frac{\gamma a^3}{\epsilon_{effM,c} R^3}\right) E'_0 e^{i\nu_c t} + 4K \frac{\wp_{ca} \rho_{ac} \gamma a^3}{\epsilon_{effQ,c} \epsilon_{effM,c} R^6} \\
&= E_{c,eff} + 4K \frac{\wp_{ca} \rho_{ac} \gamma a^3}{\epsilon_{effQ,c} \epsilon_{effM,c} R^6} \tag{3.22}
\end{aligned}$$

where $E_{c,eff}$ is the effective control field.

From the first term of eq. (3.21) and eq. (3.22) we see that in the presence of MP, the probe field and control field are modified giving the effective probe field $E_{p,eff}$ and

effective control field $E_{c,eff}$. For $E_{QS,p}$ it is modified the factor $1 + 2\frac{\gamma a^3}{\epsilon_{effM,p}R^3}$ and for $E_{QS,c}$ it is modified by a factor of $1 + 2\frac{\gamma a^3}{\epsilon_{effM,c}R^3}$. Thus, the strength of probe field depends on 2 factors, plasmonic enhancement, γ and also the QS-MP distance, R . Since the effective probe field and effective control field strength is proportional to the inverse cube root of the QS-MP distance, $\sim \frac{1}{R^3}$, thus, the field strength is very sensitive to the QS-MP distance. Besides, the effective field also depends on the plasmonic enhancement, γ . Recall that $\gamma = \frac{\epsilon_m(\omega) - \epsilon_b}{\epsilon_m(\omega) + 2\epsilon_b}$, thus the effective field strength can also be enhanced through $\epsilon_m(\omega)$. $\epsilon_m(\omega)$ depends on the laser frequency and also on the type of MP used.

Furthermore, the local field also depends on another term. For $E_{QS,p}$, this term is $4K\frac{\rho_{ba}\rho_{ab}\gamma a^3\hat{x}}{\epsilon_{effM,p}\epsilon_{effQ,p}R^6}$ and for $E_{QS,c}$, this term is $4K\frac{\rho_{ca}\rho_{ac}\gamma a^3}{\epsilon_{effQ,c}\epsilon_{effM,c}R^6}$. Thus, the local field strength depends on the plasmonic enhancement γ and the QS-MP distance R and the coherence terms ρ_{ab} and ρ_{ac} . The field strength is also proportional to $\frac{1}{R^6}$, again indicating that the local field is very sensitive to QS-MP distance. There is also a linear dependance on γ and the coherence terms. The coherence terms can be found through solving the equations of motion for the QS density matrix.

To gain further insight, we can substitute eq. (3.6), eq. (3.7), eq. (3.11) and eq. (3.12) into eq. (3.2) and eq. (3.3) and rewrite:

$$\begin{aligned}
\mathbf{E}_{QS,p} &= E_0 e^{iv_p t} \hat{e} + \frac{1}{4\pi\epsilon_0} \frac{4\pi\epsilon_0\gamma a^3 \left(E_0 e^{iv_p t} \hat{e} + \frac{1}{4\pi\epsilon_0} \frac{\mu(\rho_{12} + \rho_{21})\hat{s}}{\epsilon_{effQ,p}R^3} \right) \hat{s}}{\epsilon_{effM,p}R^3} \\
&= E_0 e^{iv_p t} \hat{e} + \frac{\gamma a^3 E_0 e^{iv_p t} \hat{s} \hat{e} + \frac{\gamma a^3 \hat{s}^2}{4\pi\epsilon_0 \epsilon_{effQ,p} R^3} \mu (\rho_{ab} + \rho_{ba})}{\epsilon_{effM,p} R^3} \\
&= E_0 e^{iv_p t} \hat{e} \left(1 + \frac{\gamma a^3 \hat{s}}{\epsilon_{effM,p} R^3} \right) + \frac{\gamma a^3 \hat{s}^2 \mu^2}{4\pi\epsilon_0 \epsilon_{effQ,p} \epsilon_{effM,p} R^6} \frac{1}{\mu} (\rho_{ab} + \rho_{ba}) \\
&= \frac{2\hbar}{\mu E_0} \Omega_p E_0 e^{iv_p t} \hat{e} + \frac{G_p}{\mu} (\rho_{ab} + \rho_{ba}) \tag{3.23}
\end{aligned}$$

$$\begin{aligned}
\mathbf{E}_{QS,c} &= E'_0 e^{iv_{ct}} \hat{e} + \frac{1}{4\pi\epsilon_0} \frac{4\pi\epsilon_0 \gamma a^3 \left(E'_0 e^{iv_{ct}} \hat{e} + \frac{1}{4\pi\epsilon_0} \frac{p_{QS,c} s \alpha}{\epsilon_{effQ,c} R^3} \right) \hat{s}}{\epsilon_{effM,c} R^3} \\
&= E'_0 e^{iv_{ct}} \hat{e} + \frac{\gamma a^3 E'_0 e^{iv_{ct}} \hat{s} \hat{e} + \frac{\gamma a^3 s^2}{4\pi\epsilon_0 \epsilon_{effQ,c} R^3} \mu (\rho_{ac} + \rho_{ca})}{\epsilon_{effM,c} R^3} \\
&= E'_0 e^{iv_{ct}} \hat{e} \left(1 + \frac{\gamma a^3 \hat{s}}{\epsilon_{effM,c} R^3} \right) + \frac{\gamma a^3 s^2 \mu^2}{4\pi\epsilon_0 \epsilon_{effQ,c} \epsilon_{effM,c} R^6} \frac{1}{\mu} (\rho_{ac} + \rho_{ca}) \\
&= \frac{2\hbar}{\mu E_0} \Omega_c E'_0 e^{iv_{ct}} \hat{e} + \frac{G_c}{\mu} (\rho_{ac} + \rho_{ca}) \tag{3.24}
\end{aligned}$$

where $G_p = \frac{\gamma a^3 s^2 \mu^2}{4\pi\epsilon_0 \epsilon_{effQ,p} \epsilon_{effM,p} R^6}$, $\Omega_p = \frac{\mu E_0}{2\hbar} \left(1 + \frac{\gamma a^3 \hat{s}}{\epsilon_{effM,p} R^3} \right)$, $G_c = \frac{\gamma a^3 s^2 \mu^2}{4\pi\epsilon_0 \epsilon_{effQ,c} \epsilon_{effM,c} R^6}$ and $\Omega_c = \frac{\mu E_0}{2\hbar} \left(1 + \frac{\gamma a^3 \hat{s}}{\epsilon_{effM,c} R^3} \right)$. The term G_p arises when the applied field polarizes the QS which in turn polarizes the MP, which then polarizes QS again and so on. Thus, the term G_p can be thought of as the self interaction of QS. As for the Ω_p term, the first term is the direct coupling of QS to the field, while the second term is the field from the MP that is induced by the applied field. G_c and Ω_c have similar interpretation.

CHAPTER 4

VECTORIAL LOCAL FIELD

4.1 Vectorial Local Field Equations For QS and MP

In the previous section, we consider the near field ($R \ll \frac{1}{k}$) for one dimension. Here we consider the exact local field for any arbitrary R . The local field of the MP has three components; the field that is due to the probe field, control field and the dipole of QS.

The field produced by a dipole is given in (Jackson, 1998):

$$\begin{aligned} E_{dip} &= \frac{1}{4\pi\epsilon_0\epsilon_{eff}}\mathbf{f}(R) \\ &= \frac{K}{\epsilon_{eff}}\mathbf{f}(R) \end{aligned} \quad (4.1)$$

where $K = \frac{1}{4\pi\epsilon_0}$ and

$$\mathbf{f}(R) = \frac{k^2}{R} (\hat{\mathbf{n}} \times \mathbf{p}) \times \hat{\mathbf{n}} + [3\hat{\mathbf{n}}(\hat{\mathbf{n}} \cdot \mathbf{p}) - \mathbf{p}] \left(\frac{1}{R^3} - \frac{ik}{R^2} \right) \quad (4.2)$$

Using vector triple product identity $(\mathbf{a} \times \mathbf{b}) \times \mathbf{c} = (\mathbf{c} \cdot \mathbf{a})\mathbf{b} - (\mathbf{c} \cdot \mathbf{b})\mathbf{a}$, then:

$$\begin{aligned} \mathbf{f}(R) &= \frac{k^2}{R} [(\hat{\mathbf{n}} \cdot \hat{\mathbf{n}})\mathbf{p} - (\hat{\mathbf{n}} \cdot \mathbf{p})\hat{\mathbf{n}}] + [3\hat{\mathbf{n}}(\hat{\mathbf{n}} \cdot \mathbf{p}) - \mathbf{p}] \left(\frac{1}{R^3} - \frac{ik}{R^2} \right) \\ &= \frac{k^2}{R} [\mathbf{p} - (\hat{\mathbf{n}} \cdot \mathbf{p})\hat{\mathbf{n}}] + [3\hat{\mathbf{n}}(\hat{\mathbf{n}} \cdot \mathbf{p}) - \mathbf{p}] \left(\frac{1}{R^3} - \frac{ik}{R^2} \right) \\ &= \mathbf{p} \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right) + (\mathbf{p} \cdot \hat{\mathbf{n}})\hat{\mathbf{n}} \left(-\frac{k^2}{R} + \frac{3}{R^3} - \frac{3ik}{R^2} \right) \\ &= A(k, R) \begin{pmatrix} p_x \\ p_y \end{pmatrix} + B(k, R) \begin{pmatrix} n_x \\ n_y \end{pmatrix} \mathbf{p} \cdot \hat{\mathbf{n}} \end{aligned} \quad (4.3)$$

where $\hat{\mathbf{n}}$ is the direction of the field due to polarization and

$$A(k, R) = \frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \quad (4.4)$$

$$B(k, R) = -\frac{k^2}{R} + \frac{3}{R^3} - \frac{3ik}{R^2} \quad (4.5)$$

$$\mathbf{p} \cdot \hat{\mathbf{n}} = p_x n_x + p_y n_y \quad (4.6)$$

In the QS dipole field, \mathbf{p} is replaced by the transition dipole moment operator $\wp_{ba} \tilde{\rho}_{ab}$ for the probe field and $\wp_{ca} \tilde{\rho}_{ac}$ for the control field:

$$\mathbf{f}_p(R) = \tilde{\rho}_{ab} \left[A(k, R) \begin{pmatrix} \wp_{ba,x} \\ \wp_{ba,y} \end{pmatrix} + B(k, R) \begin{pmatrix} n_x \\ n_y \end{pmatrix} \wp_{ba} \cdot \hat{\mathbf{n}} \right] \quad (4.7)$$

$$\mathbf{f}_c(R) = \tilde{\rho}_{ac} \left[A(k, R) \begin{pmatrix} \wp_{ca,x} \\ \wp_{ca,y} \end{pmatrix} + B(k, R) \begin{pmatrix} n_x \\ n_y \end{pmatrix} \wp_{ca} \cdot \hat{\mathbf{n}} \right] \quad (4.8)$$

where $\wp^* \cdot \hat{\mathbf{n}}' = \wp_x^* \hat{n}'_x + \wp_y^* \hat{n}'_y$.

The dipole field as derived is given by:

$$\begin{aligned} \mathbf{E}_{dip}^{QS} &= \frac{1}{4\pi\epsilon_0\epsilon_{effM}} \mathbf{f}(R) \\ &= \frac{K}{\epsilon_{effM}} \mathbf{f}(R) \end{aligned} \quad (4.9)$$

We can separate the contribution from the probe field and control field part:

$$\mathbf{E}_{dip,p}^{QS} = \frac{K}{\epsilon_{effM,p}} \mathbf{f}_p(R) \quad (4.10)$$

$$\mathbf{E}_{dip,c}^{QS} = \frac{K}{\epsilon_{effM,c}} \mathbf{f}_c(R) \quad (4.11)$$

As in the previous chapter, we write the local field separately for the contribution of probe and control field:

$$\begin{aligned}
\mathbf{E}_{MPp} &= \mathbf{E}_p + \mathbf{E}_{dip,p}^{QS} \\
&= \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{iv_p t} + \frac{K}{\epsilon_{effQ,p}} \tilde{\rho}_{ab} \left[A \begin{pmatrix} \wp_{ba,x} \\ \wp_{ba,y} \end{pmatrix} + B \begin{pmatrix} n'_x \\ n'_y \end{pmatrix} \wp_{ba} \cdot \hat{n}' \right] \quad (4.12)
\end{aligned}$$

$$\begin{aligned}
\mathbf{E}_{MPc} &= \mathbf{E}_c + \mathbf{E}_{dip,c}^{QS} \\
&= \begin{pmatrix} E'_x \\ E'_y \end{pmatrix} e^{iv_c t} + \frac{K}{\epsilon_{effQ,c}} \tilde{\rho}_{ac} \left[A \begin{pmatrix} \wp_{ca,x} \\ \wp_{ca,y} \end{pmatrix} + B \begin{pmatrix} n'_x \\ n'_y \end{pmatrix} \wp_{ca} \cdot \hat{n}' \right] \quad (4.13)
\end{aligned}$$

Recall that $p_{MP,p} = \wp_{ba} \rho_{ab}$ and $p_{MP,c} = \wp_{ca} \rho_{ac}$. Similarly, we write separately the local field of QS in terms of the contribution from probe field and control field:

$$\begin{aligned}
\mathbf{E}_{QSp} &= \mathbf{E}_p + \mathbf{E}_{dip,p}^{MP} \\
&= \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{iv_p t} + \frac{K}{\epsilon_{effM,p}} \left[A \begin{pmatrix} p_x^{MP} \\ p_y^{MP} \end{pmatrix} + B \begin{pmatrix} n_x \\ n_y \end{pmatrix} \mathbf{p}_{MP} \cdot \hat{n} \right] \\
&= \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{iv_p t} + \frac{K}{\epsilon_{effM,p}} \left[A \begin{pmatrix} \alpha_{MP} E_{MPp,x} \\ \alpha_{MP} E_{MPp,y} \end{pmatrix} + B \begin{pmatrix} n_x \\ n_y \end{pmatrix} (\alpha_{MP} E_{MPp,x} n_x + \alpha_{MP} E_{MPp,y} n_y) \right] \\
&= \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{iv_p t} + \frac{K}{\epsilon_{effM,p}} \alpha_{MP} \begin{pmatrix} A E_{MPp,x} + B E_{MPp,x} n_x^2 + B E_{MPp,y} n_x n_y \\ A E_{MPp,y} + B E_{MPp,x} n_x n_y + B E_{MPp,y} n_y^2 \end{pmatrix} \\
&= \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{iv_p t} + \frac{K}{\epsilon_{effM,p}} \alpha_{MPM} \begin{pmatrix} E_{MPp,x} \\ E_{MPp,y} \end{pmatrix} \quad (4.14)
\end{aligned}$$

$$\begin{aligned}
\mathbf{E}_{QSc} &= \mathbf{E}_c + \mathbf{E}_{dip,c}^{MP} \\
&= \begin{pmatrix} E'_x \\ E'_y \end{pmatrix} e^{iv_c t} + \frac{K}{\epsilon_{effM,c}} \alpha_{MPM} \begin{pmatrix} E_{MPc,x} \\ E_{MPc,y} \end{pmatrix} \quad (4.15)
\end{aligned}$$

where $M = \begin{pmatrix} A + Bn_x^2 & Bn_x n_y \\ Bn_x n_y & A + Bn_y^2 \end{pmatrix}$. Substituting eq. (4.12) to eq. (4.14):

$$\begin{aligned}
E_{QSp} &= \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{iv_p t} + \frac{K}{\varepsilon_{effM,p}} \alpha_{MPM} \begin{pmatrix} E_{MPp,x} \\ E_{MPp,y} \end{pmatrix} \\
&= \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{iv_p t} + \frac{K}{\varepsilon_{effM,p}} \alpha_{MPM} \left\{ \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{iv_p t} + \frac{K}{\varepsilon_{effQ,p}} \tilde{\rho}_{ab} \left[A \begin{pmatrix} \wp_{ba,x} \\ \wp_{ba,y} \end{pmatrix} + B \begin{pmatrix} n'_x \\ n'_y \end{pmatrix} \wp_{ba \cdot \hat{n}'} \right] \right\} \\
&= \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{iv_p t} + \frac{K}{\varepsilon_{effM,p}} \alpha_{MPM} \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{iv_p t} \\
&\quad + \frac{K^2}{\varepsilon_{effQ,p} \varepsilon_{effM,p}} \alpha_{MPM} \tilde{\rho}_{ab} \left[A \begin{pmatrix} \wp_{ba,x} \\ \wp_{ba,y} \end{pmatrix} + B \begin{pmatrix} n'_x \\ n'_y \end{pmatrix} \wp_{ba \cdot \hat{n}'} \right] \\
&= \left(1 + \frac{K}{\varepsilon_{effM,p}} \alpha_{MPM} \right) \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{iv_p t} \\
&\quad + \frac{K^2}{\varepsilon_{effQ,p} \varepsilon_{effM,p}} \alpha_{MPM} \tilde{\rho}_{ab} \left[A \begin{pmatrix} \wp_{ba,x} \\ \wp_{ba,y} \end{pmatrix} + B \begin{pmatrix} n'_x \\ n'_y \end{pmatrix} \wp_{ba \cdot \hat{n}'} \right] \tag{4.16}
\end{aligned}$$

$$\begin{aligned}
E_{QSc} &= \begin{pmatrix} E'_x \\ E'_y \end{pmatrix} e^{iv_c t} + \frac{K}{\varepsilon_{effM,c}} \alpha_{MPM} \left\{ \begin{pmatrix} E'_x \\ E'_y \end{pmatrix} e^{iv_c t} + \frac{K}{\varepsilon_{effQ,c}} \tilde{\rho}_{ac} \left[A \begin{pmatrix} \wp_{ca,x} \\ \wp_{ca,y} \end{pmatrix} + B \begin{pmatrix} n'_x \\ n'_y \end{pmatrix} \wp_{ca \cdot \hat{n}'} \right] \right\} \\
&= \begin{pmatrix} E'_x \\ E'_y \end{pmatrix} e^{iv_c t} + \frac{K}{\varepsilon_{effM,c}} \alpha_{MPM} \begin{pmatrix} E'_x \\ E'_y \end{pmatrix} e^{iv_c t} \\
&\quad + \frac{K^2}{\varepsilon_{effM,c} \varepsilon_{effQ,c}} \alpha_{MPM} \tilde{\rho}_{ac} \left[A \begin{pmatrix} \wp_{ca,x} \\ \wp_{ca,y} \end{pmatrix} + B \begin{pmatrix} n'_x \\ n'_y \end{pmatrix} \wp_{ca \cdot \hat{n}'} \right] \\
&= \left(1 + \frac{K}{\varepsilon_{effM,c}} \alpha_{MPM} \right) \begin{pmatrix} E'_x \\ E'_y \end{pmatrix} e^{iv_c t} \\
&\quad + \frac{K^2}{\varepsilon_{effM,c} \varepsilon_{effQ,c}} \alpha_{MPM} \tilde{\rho}_{ac} \left[A \begin{pmatrix} \wp_{ca,x} \\ \wp_{ca,y} \end{pmatrix} + B \begin{pmatrix} n'_x \\ n'_y \end{pmatrix} \wp_{ca \cdot \hat{n}'} \right] \tag{4.17}
\end{aligned}$$

where $\varepsilon_{effQ,c} = \frac{2\varepsilon_b + \varepsilon_{QS,c}}{3\varepsilon_b}$ and $\varepsilon_{QS} = 1 + \frac{\eta}{\Omega} \tilde{\rho}_{ab}$, and $\wp_{ba \cdot \hat{n}'} = \wp_{ba,x} n'_x + \wp_{ba,y} n'_y$, $\wp_{ca \cdot \hat{n}'} = \wp_{ca,x} n'_x + \wp_{ca,y} n'_y$.

4.1.1 Vectorial Local Fields in 2-dimension

Recall that $M = \begin{pmatrix} A + Bn_x^2 & Bn_x n_y \\ Bn_x n_y & A + Bn_y^2 \end{pmatrix}$ and assume that MP and QS are aligned on the x-axis as shown in Fig. 2.1, then $n_x = -1$, $n'_x = 1$, $n_y = n'_y = 0$, then $M = \begin{pmatrix} A + B & 0 \\ 0 & A \end{pmatrix}$, $\wp_{ba \cdot \hat{n}'} = \wp_{ba,x}$, $\wp_{ca \cdot \hat{n}'} = \wp_{ca,x}$. Thus in this case, eq. (4.16) and eq. (4.17) can be written as:

$$\begin{aligned}
E_{QSp} &= \begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{ivpt} + \frac{K}{\varepsilon_{effM,p}} \alpha_{MP} \begin{pmatrix} (A+B)E_x \\ AE_y \end{pmatrix} e^{ivpt} \\
&+ \frac{K^2}{\varepsilon_{effQ,p}\varepsilon_{effM,p}} \alpha_{MP} \tilde{\rho}_{ab} \left[A \begin{pmatrix} (A+B)\wp_{ba,x} \\ A\wp_{ba,y} \end{pmatrix} + B \begin{pmatrix} A+B \\ 0 \end{pmatrix} \wp_{ba,x} \right] \quad (4.18)
\end{aligned}$$

$$\begin{aligned}
E_{QSc} &= \begin{pmatrix} E'_x \\ E'_y \end{pmatrix} e^{ivct} + \frac{K}{\varepsilon_{effM,c}} \alpha_{MP} \begin{pmatrix} (A+B)E'_x \\ AE'_y \end{pmatrix} e^{ivct} \\
&+ \frac{K^2}{\varepsilon_{effM,c}\varepsilon_{effQ,c}} \alpha_{MP} \tilde{\rho}_{ac} \left[A \begin{pmatrix} (A+B)\wp_{ca,x} \\ A\wp_{ca,y} \end{pmatrix} + B \begin{pmatrix} A+B \\ 0 \end{pmatrix} \wp_{ca,x} \right] \quad (4.19)
\end{aligned}$$

Recall that $A = \frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3}$, $B = -\frac{k^2}{R} + \frac{3}{R^3} - \frac{3ik}{R^2}$, $A+B = \frac{2}{R^3} - \frac{2ik}{R^2}$ and $\alpha_{MP} = \frac{1}{K}\gamma a^3$, then eq. (4.16) and eq. (4.17) can be written in terms of its x and y components:

$$\begin{aligned}
E_{QSp,x} &= E_x e^{ivpt} + \frac{K}{\varepsilon_{effM,p}} \alpha_{MP} (A+B) E_x e^{ivpt} \\
&+ \frac{K^2}{\varepsilon_{effQ,p}\varepsilon_{effM,p}} \alpha_{MP} \tilde{\rho}_{ab} [A(A+B)\wp_{ba,x} + B(A+B)\wp_{ba,x}] \\
&= E_x e^{ivpt} \left[1 + \frac{2}{\varepsilon_{effM,p}} \gamma a^3 \left(\frac{1}{R^3} - \frac{ik}{R^2} \right) \right] \\
&+ \frac{K}{\varepsilon_{effQ,p}\varepsilon_{effM,p}} \gamma a^3 \tilde{\rho}_{ab} \left(\frac{2}{R^3} - \frac{2ik}{R^2} \right)^2 \wp_{ba,x} \\
&= E_{eff,x} + \frac{K}{\varepsilon_{effQ,p}\varepsilon_{effM,p}} \gamma a^3 \tilde{\rho}_{ab} \left(\frac{2}{R^3} - \frac{2ik}{R^2} \right)^2 \wp_{ba,x} \quad (4.20)
\end{aligned}$$

$$\begin{aligned}
E_{QSp,y} &= E_y e^{ivpt} + \frac{K}{\varepsilon_{effM,p}} \alpha_{MP} A E_y e^{ivpt} + \frac{K^2}{\varepsilon_{effQ,p}\varepsilon_{effM,p}} \alpha_{MP} \tilde{\rho}_{ab} A^2 \wp_{ba,y} \\
&= E_y e^{ivpt} \left[1 + \frac{1}{\varepsilon_{effM,p}} \gamma a^3 \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right) \right] \\
&+ \frac{K}{\varepsilon_{effQ,p}\varepsilon_{effM,p}} \gamma a^3 \tilde{\rho}_{ab} \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right)^2 \wp_{ba,y} \\
&= E_{eff,y} + \frac{K}{\varepsilon_{effQ,p}\varepsilon_{effM,p}} \gamma a^3 \tilde{\rho}_{ab} \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right)^2 \wp_{ba,y} \quad (4.21)
\end{aligned}$$

$$\begin{aligned}
E_{QSc,x} &= E'_x e^{iv_c t} + \frac{K}{\epsilon_{effM,c}} \alpha_{MP} (A+B) E'_x e^{iv_c t} \\
&\quad + \frac{K^2}{\epsilon_{effM,c} \epsilon_{effQ,c}} \alpha_{MP} \tilde{\rho}_{ac} [A(A+B) \wp_{ca,x} + B(A+B) \wp_{ca,x}] \\
&= E'_x e^{iv_c t} \left[1 + \frac{1}{\epsilon_{effM,c}} \gamma a^3 \left(\frac{2}{R^3} - \frac{2ik}{R^2} \right) \right] \\
&\quad + \frac{K}{\epsilon_{effM,c} \epsilon_{effQ,c}} \gamma a^3 \tilde{\rho}_{ac} \left(\frac{2}{R^3} - \frac{2ik}{R^2} \right)^2 \wp_{ca,x} \\
&= E'_{eff,x} + \frac{K}{\epsilon_{effM,c} \epsilon_{effQ,c}} \gamma a^3 \tilde{\rho}_{ac} \left(\frac{2}{R^3} - \frac{2ik}{R^2} \right)^2 \wp_{ca,x} \tag{4.22}
\end{aligned}$$

$$\begin{aligned}
E_{QSc,y} &= E'_y e^{iv_c t} + \frac{K}{\epsilon_{effM,c}} \alpha_{MP} A E'_y e^{iv_c t} + \frac{K^2}{\epsilon_{effM,c} \epsilon_{effQ,c}} \alpha_{MP} \tilde{\rho}_{ac} A^2 \wp_{ca,y} \\
&= E'_y e^{iv_c t} \left[1 + \frac{1}{\epsilon_{effM,c}} \gamma a^3 \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right) \right] \\
&\quad + \frac{K}{\epsilon_{effM,c} \epsilon_{effQ,c}} \gamma a^3 \tilde{\rho}_{ac} \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right)^2 \wp_{ca,y} \\
&= E'_{eff,y} + \frac{K}{\epsilon_{effM,c} \epsilon_{effQ,c}} \gamma a^3 \tilde{\rho}_{ac} \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right)^2 \wp_{ca,y} \tag{4.23}
\end{aligned}$$

where $E_{eff,x} = E_x e^{iv_p t} \left[1 + \frac{2}{\epsilon_{effM,p}} \gamma a^3 \left(\frac{1}{R^3} - \frac{ik}{R^2} \right) \right]$,
 $E_{eff,y} = E_y e^{iv_p t} \left[1 + \frac{1}{\epsilon_{effM,p}} \gamma a^3 \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right) \right]$,
 $E'_{eff,x} = E'_x e^{iv_c t} \left[1 + \frac{1}{\epsilon_{effM,c}} \gamma a^3 \left(\frac{2}{R^3} - \frac{2ik}{R^2} \right) \right]$ and
 $E'_{eff,y} = E'_y e^{iv_c t} \left[1 + \frac{1}{\epsilon_{effM,c}} \gamma a^3 \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right) \right]$ are the effective probe field in x and y-component and the effective control field in the x and y-component.

Instead of looking at the QS-MP system as a one-dimensional problem as we did in the previous section, here we derive a general vectorial description for the QS-MP system in two-dimensional in which we do not specify the probe and control laser direction. As in the linear case, we see the probe and control field are modified in the presence of MP. The probe and control field also have a linear dependence on the plasmonic enhancement, γ , similar as in the linear model. However, the effective probe and control field dependence on the QS-MP distance is now different. In the x-component for both effective probe and control field, there is still an inverse cube root dependence on the QS-MP distance, $\sim \frac{1}{R^3}$,

however, there is also an additional competing effect from the negative inverse square root dependence on QS-MP distance $\sim -\frac{1}{R^2}$. On the other hand, in the y-component for both effective probe and control field, the leading term in the dependence on QS-MP distance is $\sim -\frac{1}{R^3}$, thus, decreasing QS-MP distance actually diminishes the effective probe and control field strength.

The local field of QS also depends on another term. The local field strength depends linearly on the plasmonic enhancement γ and coherence terms $\tilde{\rho}_{ab}$ and $\tilde{\rho}_{ac}$ and nonlinearly on the QS-MP distance with the leading term of $\frac{1}{R^6}$.

Since $\hbar\Omega_b = \wp_{ab} \cdot \mathbf{E}_{QSp}$ and $\hbar\Omega_c = \wp_{ac} \cdot \mathbf{E}_{QSc}$ then:

$$\begin{aligned}
\hbar\Omega_{ab} &= \wp_{ab} \begin{pmatrix} E_{QSp,x} \\ E_{QSp,y} \end{pmatrix} \\
&= \wp_{ab} \left[\begin{pmatrix} E_x \\ E_y \end{pmatrix} e^{i\nu_p t} + \frac{1}{\epsilon_{effQ,p}} \left(\mathbf{P} e^{i\nu_p t} + \frac{\tilde{\rho}_{ab}}{\epsilon_{effM,p}} \mathbf{R} \right) \right] \\
&= \hbar\Omega_b e^{i\nu_p t} + \frac{1}{\epsilon_{effQ,p}} \left(\wp_{ab} \cdot \mathbf{P} e^{i\nu_p t} + \frac{\tilde{\rho}_{ab}}{\epsilon_{effM,p}} \wp_{ab} \cdot \mathbf{R} \right) \\
&= \hbar\Omega_b e^{i\nu_p t} + \frac{1}{\epsilon_{effQ,p}} \wp_{ab} \cdot \mathbf{P} e^{i\nu_p t} + \frac{1}{\epsilon_{effM,p}} \frac{3\epsilon_b \tilde{\rho}_{ab}}{2\epsilon_b + 1 + \frac{\eta_p}{\Omega_{ab}} \tilde{\rho}_{ab}} \wp_{ab} \cdot \mathbf{R} \quad (4.24)
\end{aligned}$$

$$\begin{aligned}
\hbar\Omega_{ac} &= \wp_{ac} \begin{pmatrix} E_{QSc,x} \\ E_{QSc,y} \end{pmatrix} \\
&= \wp_{ac} \left[\begin{pmatrix} E'_x \\ E'_y \end{pmatrix} e^{i\nu_c t} + \frac{1}{\epsilon_{effQ,c}} \left(\mathbf{Q} e^{i\nu_c t} + \frac{\tilde{\rho}_{ac}}{\epsilon_{effM,c}} \mathbf{S} \right) \right] \\
&= \hbar\Omega_c e^{i\nu_c t} + \frac{1}{\epsilon_{effQ,c}} \left(\wp_{ac} \cdot \mathbf{Q} e^{i\nu_c t} + \frac{\tilde{\rho}_{ac}}{\epsilon_{effM,c}} \wp_{ac} \cdot \mathbf{S} \right) \\
&= \hbar\Omega_c e^{i\nu_c t} + \frac{1}{\epsilon_{effQ,c}} \wp_{ac} \cdot \mathbf{Q} e^{i\nu_c t} + \frac{1}{\epsilon_{effM,c}} \frac{3\epsilon_b \tilde{\rho}_{ac}}{2\epsilon_b + 1 + \frac{\eta_c}{\Omega_{ac}} \tilde{\rho}_{ac}} \wp_{ac} \cdot \mathbf{S} \quad (4.25)
\end{aligned}$$

where $\eta_c = \frac{N|\wp_{ac}|^2}{\hbar\epsilon_0}$ and $\eta_p = \frac{N|\wp_{ab}|^2}{\hbar\epsilon_0}$. Ω_{ab} and Ω_{ac} are the effective Rabi frequencies of the QS when the QS is coupled to the metallic nanoparticle. We see that the Rabi frequencies are modified by the 2nd and 3rd term in eq. (4.24) and eq. (4.25). If we let R tends to infinity, then we see that we recover the Rabi frequencies of the EIT system.

The three-level EIT scheme equations are given by:

$$\begin{aligned}
\frac{\partial \rho_{aa}}{\partial t} &= -2(\gamma_{ab} + \gamma_{ac})\rho_{aa} + i\Omega_d \rho_{ba} - i\Omega_d^* \rho_{ab} + i\Omega_p \rho_{ca} - i\Omega_p^* \rho_{ac} \\
\frac{\partial \rho_{ab}}{\partial t} &= -(\gamma_{ab} + \gamma_{ac})\rho_{ab} - i\Omega_d(\rho_{aa} - \rho_{bb}) + i\Omega_p \rho_{cb} \\
\frac{\partial \rho_{ac}}{\partial t} &= -(\gamma_{ab} + \gamma_{ac} + i\Delta)\rho_{ac} + i\Omega_d \rho_{bc} - i\Omega_p(\rho_{aa} - \rho_{cc}) \\
\frac{\partial \rho_{bb}}{\partial t} &= 2\gamma_{ab}\rho_{aa} - i\Omega_d \rho_{ba} + i\Omega_d^* \rho_{ab} \\
\frac{\partial \rho_{bc}}{\partial t} &= -(\Gamma_{bc} + i\Delta)\rho_{bc} + i\Omega_d^* \rho_{ac} - i\Omega_p \rho_{ba} \\
\frac{\partial \rho_{cc}}{\partial t} &= 2\gamma_{ac}\rho_{aa} - i\Omega_p \rho_{ca} + i\Omega_p^* \rho_{ac}
\end{aligned} \tag{4.26}$$

where $2\gamma_{ij}$ are the decay rates from $|i\rangle$ to $|j\rangle$ and Γ_{bc} is the dephasing rate of the Raman coherence, $\Omega_d = \frac{\mathcal{E}_{ab}\xi_p}{\hbar}$ and $\Omega_p = \frac{\mathcal{E}_{ac}\xi_c}{\hbar}$.

For the three-level Raman scheme, the steady-state density matrix elements are given by:

$$\tilde{\rho}_{ac} = \frac{1}{\Upsilon_{ac}} \{-i\Omega_{ac} w_{ac} + i\Omega_b \tilde{\rho}_{bc}\} \tag{4.27}$$

$$\tilde{\rho}_{ba} = \frac{1}{\Upsilon_{ab}^*(\omega)} \{i\Omega_b^* w_{ab} - i\Omega_{ac}^* \tilde{\rho}_{bc}\} \tag{4.28}$$

$$\tilde{\rho}_{bc} = \frac{1}{\Upsilon_{bc}(\omega)} \{i\Omega_b^* \tilde{\rho}_{ac} - i\Omega_{ac} \tilde{\rho}_{ba}\} \tag{4.29}$$

with $\Omega_b = \Omega\eta + G\rho_{ac}$, $\Upsilon_{ac} = \gamma_{ac} - i\Delta_c$, $\Upsilon_{ab}(\omega) = \gamma_{ab} - i\Delta(\omega)$, $\Upsilon_{bc}(\omega) = \gamma_{bc} - i(\Delta_c - \Delta(\omega))$ and $w_{ab} = \rho_{aa}^{(0)} - \rho_{bb}^{(0)}$, $w_{ac} = \rho_{aa}^{(0)} - \rho_{cc}^{(0)}$. The effective decoherences are $\gamma_{ac} = \frac{\Gamma_c + \Gamma_b}{2} + \gamma_{ac}^d$, $\gamma_{ab} = \frac{\Gamma_c + \Gamma_b}{2} + \gamma_{ab}^d$ with Γ_b, Γ_c as the spontaneous emission rates, $\gamma_{ac}^d, \gamma_{ab}^d$ are dephasing rates, $\Delta(\omega) = \omega - \omega_{ab}$ and $\Delta_c = \nu_c - \omega_{ac}$ are the detunings. The radiative rate of the QS can be modified by the MP as $\Gamma_{b,c} = |f|^2 \gamma_0$ where $f = 1 + g\alpha_{MP}/\epsilon'_{eff} d^3$.

Solving eq. (4.27), (4.28) and (4.29):

$$\begin{aligned}
\tilde{\rho}_{ac} &= -i \frac{w_{ac} \Omega_{ac}^2 \Omega_{ac}^* + w_{ac} \Upsilon_{ab}^* \Upsilon_{bc} \Omega_{ac} - |\Omega_b|^2 w_{ab} \Omega_{ac}}{|\Omega_b|^2 \Upsilon_{ab}^* + \Upsilon_{ac} \Upsilon_{ab}^* \Upsilon_{bc} + \Upsilon_{ac} |\Omega_{ac}|^2} \\
\tilde{\rho}_{ba} &= i \frac{|\Omega_b|^2 \Omega_b^* w_{ab} + \Omega_b^* w_{ab} \Upsilon_{ac} \Upsilon_{bc} - \Omega_b^* w_{ac} |\Omega_{ac}|^2}{|\Omega_b|^2 \Upsilon_{ab}^* + \Upsilon_{ac} \Upsilon_{ab}^* \Upsilon_{bc} + \Upsilon_{ac} |\Omega_{ac}|^2} \\
\tilde{\rho}_{bc} &= \frac{\Omega_b^* w_{ab} \Upsilon_{ac} \Omega_{ac} + \Omega_b^* w_{ac} \Upsilon_{ab}^* \Omega_{ac}}{|\Omega_b|^2 \Upsilon_{ab}^* + \Upsilon_{ac} \Upsilon_{ab}^* \Upsilon_{bc} + \Upsilon_{ac} |\Omega_{ac}|^2}
\end{aligned} \tag{4.30}$$

Furthermore, if we assume that all atoms are initially in ground state $|b\rangle$, then $w_{ac} = 0$ and $w_{ab} = -1$ and the equations become:

$$\begin{aligned}\tilde{\rho}_{ac} &= -i \frac{|\Omega_b|^2 \Omega_{ac}}{|\Omega_b|^2 \Upsilon_{ab}^* + \Upsilon_{ac} \Upsilon_{ab}^* \Upsilon_{bc} + \Upsilon_{ac} |\Omega_{ac}|^2} \\ \tilde{\rho}_{ba} &= -i \frac{|\Omega_b|^2 \Omega_b^* + \Omega_b^* \Upsilon_{ac} \Upsilon_{bc}}{|\Omega_b|^2 \Upsilon_{ab}^* + \Upsilon_{ac} \Upsilon_{ab}^* \Upsilon_{bc} + \Upsilon_{ac} |\Omega_{ac}|^2} \\ \tilde{\rho}_{bc} &= -\frac{\Omega_b^* \Upsilon_{ac} \Omega_{ac}}{|\Omega_b|^2 \Upsilon_{ab}^* + \Upsilon_{ac} \Upsilon_{ab}^* \Upsilon_{bc} + \Upsilon_{ac} |\Omega_{ac}|^2}\end{aligned}\quad (4.31)$$

and $\tilde{\rho}_{ab} = \tilde{\rho}_{ba}^*$:

$$\tilde{\rho}_{ab} = i \Omega_b \frac{|\Omega_b|^2 + \Upsilon_{ac}^* \Upsilon_{bc}^*}{|\Omega_b|^2 \Upsilon_{ab} + \Upsilon_{ac}^* \Upsilon_{ab} \Upsilon_{bc}^* + \Upsilon_{ac}^* |\Omega_{ac}|^2}\quad (4.32)$$

Since $|\Omega_b| \ll |\Upsilon|$ we may neglect $|\Omega_b|^2$, then

$$\tilde{\rho}_{ab} = i \frac{\Omega_b \Upsilon_{bc}^*}{\Upsilon_{ab} \Upsilon_{bc}^* + I_c}\quad (4.33)$$

$$\tilde{\rho}_{ac} = -i \frac{|\Omega_b|^2 \Omega_{ac}}{\Upsilon_{ac} (\Upsilon_{ab}^* \Upsilon_{bc} + I_c)}\quad (4.34)$$

where $I_c = |\Omega_{ac}|^2$

Thus, we have derived the steady state coherences given by eq. (4.33) and eq. (4.34). However, the complex decoherences are also modified by the MP, thus we rewrite eq. (4.33) and eq. (4.34) as:

$$\tilde{\rho}_{ab} = i \frac{\Omega_b \Upsilon_{bc}^*}{\Upsilon'_{ab} \Upsilon_{bc}^* + I_c}\quad (4.35)$$

where $\Upsilon'_{ab}(\omega) = \Upsilon_{ab}(\omega) - iG$ where $G = K \frac{4|\beta|^2 \gamma a^3}{\hbar \epsilon_{eff} \epsilon'_{eff} R^6}$. Furthermore, if we include the effect of MP on $\tilde{\rho}_{ab}$ through $\epsilon_{eff Q,p} = \frac{2\epsilon_b + \epsilon_{QS}}{3\epsilon_b}$ where $\epsilon_{QS} = 1 + \frac{\eta}{\Omega} \tilde{\rho}_{ab}$. Then we rewrite eq. (4.35) as:

$$\begin{aligned}
\tilde{\rho}_{ab} &= i \frac{\Omega_b \Upsilon_{bc}^*}{(\Upsilon_{ab} - iG) \Upsilon_{bc}^* + I_c} \\
&= i \frac{\Omega_b \Upsilon_{bc}^*}{\left(\Upsilon_{ab} - iK \frac{4|\mathcal{J}|^2 \gamma a^3}{\hbar \varepsilon_{eff} \varepsilon'_{eff} R^6} \right) \Upsilon_{bc}^* + I_c} \\
&= i \frac{\Omega_b \Upsilon_{bc}^*}{\left(\Upsilon_{ab} - iK \frac{4|\mathcal{J}|^2 \gamma a^3}{\hbar \varepsilon_{eff} R^6} \frac{3\varepsilon_b \frac{\Omega}{\eta}}{\frac{\Omega}{\eta} (2\varepsilon_b + 1) + \tilde{\rho}_{ab}} \right) \Upsilon_{bc}^* + I_c} \tag{4.36}
\end{aligned}$$

Substituting Ω_b into these expressions:

$$\begin{aligned}
\tilde{\rho}_{ab} &= i(\Omega_{ab} e^{iv_p t} + \frac{1}{\hbar \varepsilon_{eff,p}} \mathcal{J}_{ab} \cdot \mathbf{P} e^{iv_p t} \\
&\quad + \frac{1}{\hbar \varepsilon_{eff,p}} \frac{3\varepsilon_b \tilde{\rho}_{ab}}{2\varepsilon_b + 1 + \frac{\eta_p}{\Omega_{ab}} \tilde{\rho}_{ab}} \mathcal{J}_{ab} \cdot \mathbf{R}) \frac{\Upsilon_{bc}^*(\omega)}{\Upsilon_{ab}(\omega) \Upsilon_{bc}^*(\omega) + I_c} \\
&= i\Omega_{ab} \frac{\Upsilon_{bc}^*(\omega)}{\Upsilon_{ab}(\omega) \Upsilon_{bc}^*(\omega) + I_c} e^{iv_p t} + i \frac{1}{\hbar \varepsilon_{eff,p}} \mathcal{J}_{ab} \cdot \mathbf{P} \frac{\Upsilon_{bc}^*(\omega)}{\Upsilon_{ab}(\omega) \Upsilon_{bc}^*(\omega) + I_c} e^{iv_p t} \\
&\quad + i \frac{1}{\hbar \varepsilon_{eff,p}} \frac{3\varepsilon_b \tilde{\rho}_{ab}}{2\varepsilon_b + 1 + \frac{\eta_p}{\Omega_{ab}} \tilde{\rho}_{ab}} \mathcal{J}_{ab} \cdot \mathbf{R} \frac{\Upsilon_{bc}^*(\omega)}{\Upsilon_{ab}(\omega) \Upsilon_{bc}^*(\omega) + I_c} \tag{4.37}
\end{aligned}$$

$$\tilde{\rho}_{ab} = \tilde{\rho}_{ab}^\infty \left[1 + \frac{1}{\Omega} \left(X + \frac{Y \tilde{\rho}_{ab}}{\kappa + \tilde{\rho}_{ab}} \right) \right] \tag{4.38}$$

where $X = \frac{1}{\varepsilon_{eff}} \mathbf{g} \cdot \mathbf{P}$, $Y = \frac{1}{\varepsilon_{eff}} g \cdot Q \frac{3\varepsilon_b \Omega}{\eta}$

CHAPTER 5

RESULTS AND DISCUSSION

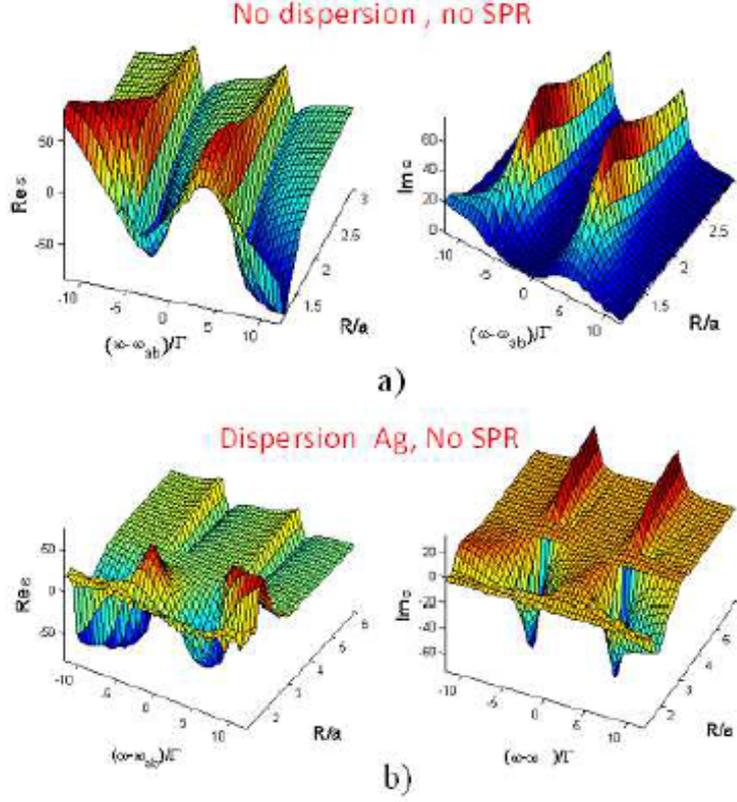


Figure 5.1: (The spectra for the real and imaginary parts of the dielectric function ϵ_{QS} versus R the spacing between the QS and MP for : a) $\epsilon_m = 0.453 + 3.35i$ with a small enhancement of $\gamma \simeq 3$. b) $\epsilon_m = -4.9 + 0.05i$ with $\gamma = 3.34 + 0.06i$ for Ag using $\omega_p = 9.1\text{eV} = 2.2 \times 10^{15}\text{s}^{-1}$, $\Gamma_m = 18\text{meV} = 2.73 \times 10^{13}\text{s}^{-1}$, $\epsilon_\infty = 3.7$ corresponding to $\omega_{SP} = 5.34 \times 10^{15}\text{s}^{-1}$, $\omega_{ab} = 2\pi c/(400\text{nm}) = 4.71 \times 10^{15}\text{s}^{-1}$. Other parameters are: $\epsilon_b = 1.5$, $\Omega_c = 5\Gamma$, the probe field amplitude $E_0 = 0.1\hbar\Gamma/\varrho$, $\varrho = 2 \times 10^{-29}\text{Cm}^{-1}$, where $\Gamma = 10^9\text{s}^{-1}$ (Folk et al., 2001) for the decay rate.

We have discussed the linear and vectorial model of the QS-MP local field. In the derivation in previous sections, we have taken into account the effect of both control and probe field. In this section, we focus on the vectorial model and the effect from probe field.

Recall that from previous chapter, we have derived the equations:

$$E_{QS,p,x} = E_x e^{i\nu p t} \left[1 + \frac{2}{\epsilon_{effM,p}} \gamma a^3 \left(\frac{1}{R^3} - \frac{ik}{R^2} \right) \right] + \frac{K}{\epsilon_{effQ,p} \epsilon_{effM,p}} \gamma a^3 \tilde{\rho}_{ab} \left(\frac{2}{R^3} - \frac{2ik}{R^2} \right)^2 \delta_{ba,x}$$

$$E_{QS,p,y} = E_y e^{i\nu p t} \left[1 + \frac{1}{\epsilon_{effM,p}} \gamma a^3 \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right) \right] + \frac{K}{\epsilon_{effQ,p} \epsilon_{effM,p}} \gamma a^3 \tilde{\rho}_{ab} \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right)^2 \delta_{ba,y}$$

The main results are obtained when the local probe field is large due to the near field or plasmonic enhancement by the MP, i.e. Ω_b is comparable to or larger than Ω_c . The Ω_b from Eq. (4.24) is substituted into the density matrix equation for the three-level system and the coherences to are numerically computed at any time. Quasi steady state solution for $\tilde{\rho}_{ab}$ is obtained for large times ($250\Gamma^{-1}$) to compute the susceptibility $\chi = \frac{N|\delta_{ab}|^2}{\hbar\epsilon_0} \frac{\tilde{\rho}_{ab}}{\Omega}$ and the dielectric function $\epsilon_{QS} = 1 + \chi$ (Ooi & Tan, 2013).

Fig. 5.1a) shows the computed dispersion spectra ($\text{Re}\epsilon_{QS}$) and absorption spectra ($\text{Im}\epsilon_{QS}$) for metal dielectric constant, $\epsilon_m = 0.453 + 3.35i$, which corresponds to a small plasmonic enhancement of $\gamma \simeq 3$. From the figure we see that for R larger than $2a$, we see the usual EIT peak. However at small separation, the EIT peaks diminish and broaden. This is due to the enhanced probe field at small separation R , which competes with the EIT effect.

The spectra in Fig. 5.1b) is computed using parameters of actual silver (Ag) nanoparticle with dispersion (Folk et al., 2001). The metal dielectric constant computed using eq. (3.10) gives the value $\epsilon_m = -4.9 + 0.05i$. Interesting feature is found for the absorption spectra. Again, we found that the EIT peaks reduces as the separation R decreases. At $R \sim 4a$, the EIT peaks are completely quenched as the enhanced probe field matches the control field, here the $\text{Im}\epsilon_{QS}$ vanishes while $\text{Re}\epsilon_{QS}$ is constant for all frequencies. At

$R \prec 4a$, we see the interesting feature in which the absorption spectra goes into the negative values, indicating that there is a gain region. In this region, Ω_b is enhanced exceeding Ω_c such that the field inside QS becomes sufficiently high to excite some population to level $|a\rangle$ creating a high gain (negative absorption).

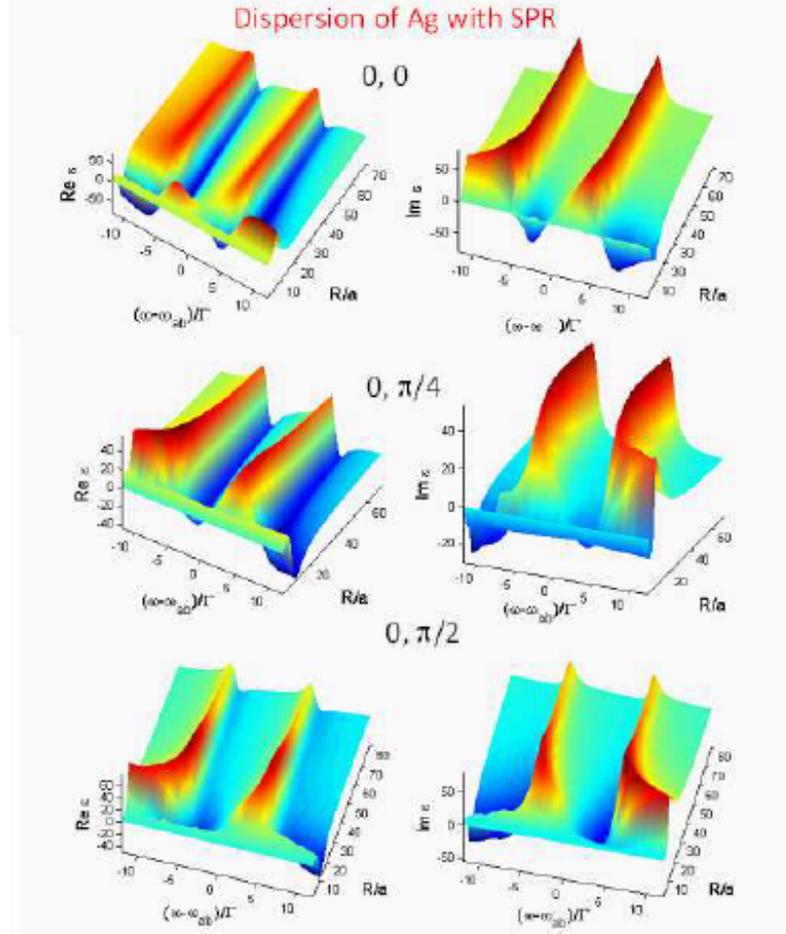


Figure 5.2: (The spectra of ϵ_{QS} versus R for different ϕ, θ with surface plasmon resonance (SPR) condition with $\omega_{ab} = \omega_{SP} = 5.34 \times 10^{15} \text{s}^{-1}$, which gives a large enhancement, $\gamma = 0.88 + 131i$ at $\omega = \omega_{SP}$. All other parameters are the same as in Fig. 5.1b.

Besides the metal dielectric function, the QS spectra is also affected by the probe field direction θ and polarization or relative phase ϕ . For the case without SPR, or $\omega_{ab} \neq \omega_{SP}$ the spectra hardly depend on θ . For $\omega_{ab} = \omega_{SP}$, the spectra are more sensitive to θ or the polarization and the relative phase ϕ of the probe laser. Thus, the spectra with SPR for different field direction and polarization is plotted in Fig. 5.2 and Fig. 5.3.

When the surface plasmon resonance (SPR) condition $\epsilon_m(\omega_{SP}) + 2\epsilon_b \simeq 0$ is satisfied, plasmonic enhancement effect occurs giving a large plasmonic enhancement, $\text{Im } \gamma \simeq 130$

($\text{Re } \gamma \simeq 0$) at $\omega = \omega_{SP}$. To see the plasmonic effects on the EIT profile, we set $\omega_{ab} = \omega_{SP}$. As shown in Fig. 5.2, the EIT peaks start to be affected by MP even at very large R . In Fig. 5.2, the spectra for different probe field direction, θ is plotted. The spectra undergoes a transition for separation smaller than $R = 50a$ and the transition is dependent on the direction and phase angles. For $\phi = 0$ with $\theta = \pi/4$ and $\pi/2$, one of the EIT peaks shifts toward higher frequencies as the separation decreases, in addition to the existence of gain region with negative $\text{Im } \epsilon_{QS}$. This frequency shift and the SPR-mediated long range Forster interaction are the unique features not found in cases without SPR enhancement.

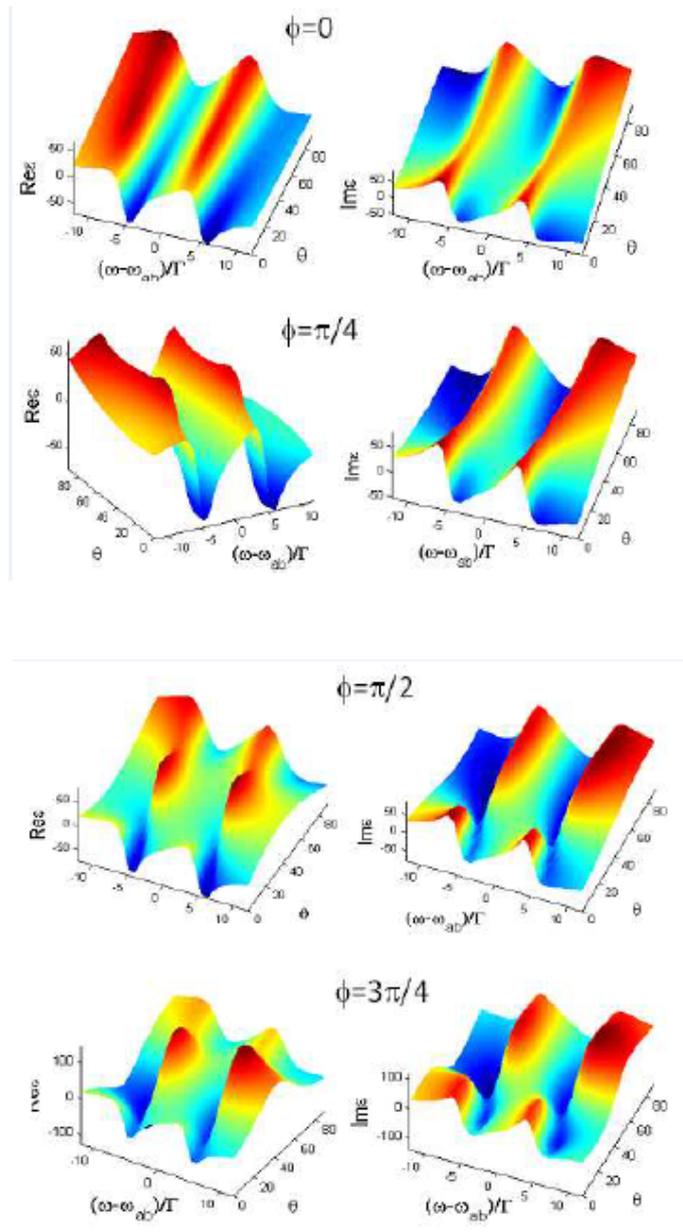


Figure 5.3: (The spectra of $\text{Re } \epsilon_{QS}$ and $\text{Im } \epsilon_{QS}$ versus θ the incident angle of the probe laser with SPR for various phase angles $\phi = 0, \pi/4, \pi/2, 3\pi/4$ at $R = 15a$. Other parameters are the same as in Fig. 5.2.

Fig. 5.3 shows the variations of the dispersion and absorption spectra with the incident angle θ and the quadrature phase ϕ at separation $R = 15a$. The ϵ_{QS} has positive and negative values that depend on the frequency and it shows small variation with θ for $\phi = 0$ and $\pi/4$. However, for $\phi = \pi/2$ and $3\pi/4$, the positions of the positive and negative peaks are shifted significantly at around $\theta = \pi/4$, i.e. when the x and y components have equal strength. This feature gives rise to directional dependence gain/absorption. These results show that the laser phase and polarization have important effects on the optical properties of the QS with nearby MP, especially in the presence of SPR.

CHAPTER 6

CONCLUSION

We have studied in details the effects of MP on a QS driven by a control laser. Analytical expressions were derived for the QS-MP system in the near field picture as well as in the vectorial picture. In the near field limit, the local field of QS is enhanced through plasmonic enhancement and the interparticle distance. The local field was found to depend on the inverse cube root of the interparticle distance and depend linearly on plasmonic enhancement. The vectorial description gives a more general picture of the problem by allowing the freedom to vary the probe field direction and polarization. In vectorial picture, similar results was found as in the near field limit. The local field depends linearly on the plasmonic enhancement in both x and y-direction and also depends on the inverse cube root of the interparticle distance in the x-direction. However in the vectorial case, it was found that there is also a competing effect coming from a negative inverse square root dependence of the local field on the interparticle distance in the x-direction. Moreover, in the y-direction, the leading term dependence of the local field on interparticle distance is negative inverse cube root.

Numerically, we focus on the result where the local field is large due to near field or plasmonic enhancement or both. The absorption and dispersion spectra are plotted for different direction, polarization and phase of the probe laser. With small plasmonic enhancement, it was found that at sufficiently small spacing between QS and MP, the electromagnetic induced transparency of the QS is quenched while the optical properties and the underlying quantum mechanisms (inferred from the spectra) of the QS are drastically altered. This is due to the near field enhancement in which the probe field is enhanced as the interparticle spacing decreases until subsequently the EIT peaks completely disappears. In the case where there is dispersion, it was found that not only that the EIT effects were quenched but there is also a negative region in the absorption spectra indicating that there is a gain region at sufficiently small spacing.

Besides the interparticle spacing, the presence of surface plasmon resonance also significantly enhances the local field of QS. With the effect of surface plasmon resonance, the QS spectra was found to be affected at an interparticle distance that is about an order magnitude larger than in the case without surface plasmon resonance. Furthermore, the absorption and dispersion spectra are plotted for different laser direction and polarization. It was found that the spectra indeed does have directional dependence and the features of the profile are more marked in the presence of surface plasmon resonance. These results show that the laser direction and phase have important effects on the optical properties of the QS with nearby MP.

By manipulating the different parameters, the features found in the dispersion/absorption spectra show that the QS-MP system could be switched between electromagnetic induced transparency, absorptive and gain/lasing regimes, providing coherent control of optical properties of nanomaterials for quantum nanophotonics devices.

Appendices

APPENDIX A

MULTIPOLE EXPANSION

It is usually difficult to evaluate exactly the potential of an arbitrary charge configuration. If the charge configuration does not exhibit considerable symmetry or if the potential is evaluated at somewhere outside of the symmetry axis, then evaluation of Coulomb's law and Biot-Savart's law become difficult. Thus, it is useful to have an approximation technique to evaluate the potential of an arbitrary charge configuration. The approximation employs the power series expansions of the potential and keeping terms up to the order of the precision desired (Griffiths, 1999).

Imagine a charge distribution with charge Q , then at a point far away from the distribution, this charge distribution is approximately a point charge, thus the potential of a point charge with the charge replaced by the total charge of the distribution will give a good approximation to the potential, $V \simeq \frac{1}{4\pi\epsilon_0} \frac{Q}{r}$, where r is the average distance of the center of the charge distribution to the point where the potential is evaluated. In fact, as will be seen later this term is the first order approximation to an arbitrary charge distribution.

But what if the charge distribution has a net charge $Q = 0$, what is the potential of this charge distribution at point far away from the distribution? If the 'point charge' approximation above is used, then the potential is zero. But this is incorrect, the potential at a point far away is very small but not zero.

As an example, solve the potential for a pair of closely spaced charges of opposite sign (an electric dipole) at a point far away from the dipole. In this case the net charge of the distribution is zero. Let the positive charge q be at $\frac{1}{2}\mathbf{d}$ from the origin and the negative charge $-q$ to be at $-\frac{1}{2}\mathbf{d}$ from the origin. The potential at \mathbf{r} due to the two charges are given by:

$$\begin{aligned}
V(\mathbf{r}) &= \frac{1}{4\pi\epsilon_0} \left(\frac{q}{|\mathbf{r} - \frac{1}{2}\mathbf{d}|} + \frac{-q}{|\mathbf{r} + \frac{1}{2}\mathbf{d}|} \right) \\
&= \frac{q}{4\pi\epsilon_0} \left(\frac{1}{|\mathbf{r} - \frac{1}{2}\mathbf{d}|} - \frac{1}{|\mathbf{r} + \frac{1}{2}\mathbf{d}|} \right)
\end{aligned} \tag{A.1}$$

Recall the law of cosines: $c^2 = a^2 + b^2 - 2\mathbf{a} \cdot \mathbf{b} = a^2 + b^2 - 2ab \cos \theta$, where θ is the angle subtended by a and b . Then the denominators in the parantheses of eq. (A.1) can be rewritten using law of cosines:

$$\begin{aligned}
V(\mathbf{r}) &= \frac{q}{4\pi\epsilon_0} \left(\frac{1}{\sqrt{r^2 + (\frac{d}{2})^2 - rd \cos \theta}} - \frac{1}{\sqrt{r^2 + (\frac{d}{2})^2 + rd \cos \theta}} \right) \\
&= \frac{q}{4\pi\epsilon_0 r} \left(\frac{1}{\sqrt{1 + (\frac{d}{2r})^2 - \frac{d}{r} \cos \theta}} - \frac{1}{\sqrt{1 + (\frac{d}{2r})^2 + \frac{d}{r} \cos \theta}} \right)
\end{aligned} \tag{A.2}$$

At point far away from the charge $r \gg d$, only first order terms in $\frac{d}{r}$ are retained:

$$V(\mathbf{r}) = \frac{q}{4\pi\epsilon_0 r} \left(\frac{1}{\sqrt{1 - \frac{d}{r} \cos \theta}} - \frac{1}{\sqrt{1 + \frac{d}{r} \cos \theta}} \right) \tag{A.3}$$

Recall the binomial series formula:

$$\begin{aligned}
(1+x)^\alpha &= \sum_{k=0}^{\infty} \binom{\alpha}{k} x^k \\
&= 1 + \alpha x + \frac{\alpha(\alpha-1)}{2!} x^2 + \frac{\alpha(\alpha-1)(\alpha-2)}{3!} x^3 + \dots
\end{aligned} \tag{A.4}$$

Rewriting the denominator of eq. (A.2):

$$\left(1 \mp \frac{d}{r} \cos \theta \right)^{-\frac{1}{2}} = 1 \pm \frac{d}{2r} \cos \theta \pm \frac{3}{8} \left(\frac{d}{r} \right)^2 \cos^2 \theta \pm \dots \tag{A.5}$$

Again keeping only first order terms in $\frac{d}{r}$ then eq. (A.3) can be written as:

$$\begin{aligned}
V(\mathbf{r}) &= \frac{q}{4\pi\epsilon_0 r} \left(1 + \frac{d}{2r} \cos \theta - 1 + \frac{d}{2r} \cos \theta \right) \\
&= \frac{qd}{4\pi\epsilon_0 r^2} \cos \theta
\end{aligned} \tag{A.6}$$

Multiply the numerator and denominator of eq. (A.6) by r and notice that $r \cdot d = rd \cos \theta$, then:

$$\begin{aligned} V(\mathbf{r}) &= \frac{\mathbf{r} \cdot (q\mathbf{d})}{4\pi\epsilon_0 r^3} \\ &= \frac{\mathbf{r} \cdot (\mathbf{p})}{4\pi\epsilon_0 r^3} \end{aligned} \quad (\text{A.7})$$

where $\mathbf{p} \equiv q\mathbf{d}$ is the dipole moment of the charge pair. This expression however is not exact as we have neglected the higher order terms and only kept the leading dipole term. We can imagine another configuration where two physical dipoles are aligned antiparallel to each other with the four charges placed at the corner of a square (quadrupole configuration). From eq. (A.7) then we can see that for large r , the dipole term is approximately zero, thus its potential is dominated by the next term in the expansion, the quadrupole term.

Now, we would like to find a general power series expansion for an arbitrary charge distribution. Let \mathbf{r} be the point that the potential is evaluated and \mathbf{r}' to be the point of the distribution. The potential at \mathbf{r} due to an arbitrary charge distribution is given by:

$$V(\mathbf{r}) = \frac{1}{4\pi\epsilon_0} \int \frac{1}{|\mathbf{r} - \mathbf{r}'|} \rho(\mathbf{r}') d\tau' \quad (\text{A.8})$$

where ρ is the charge density and $d\tau'$ is the volume element of the charge distribution. Using law of cosines:

$$\begin{aligned} |\mathbf{r} - \mathbf{r}'|^2 &= r^2 + r'^2 - 2rr' \cos \theta' \\ &= r^2 \left(1 + \left(\frac{r'}{r}\right)^2 - 2\left(\frac{r'}{r}\right) \cos \theta' \right) \\ |\mathbf{r} - \mathbf{r}'| &= r\sqrt{1 + \epsilon} \end{aligned} \quad (\text{A.9})$$

where $\epsilon = \left(\frac{r'}{r}\right)^2 - 2\left(\frac{r'}{r}\right) \cos \theta = \left(\frac{r'}{r}\right) \left(\frac{r'}{r} - 2 \cos \theta'\right)$. Then using binomial expansion:

$$\begin{aligned}
\frac{1}{|\mathbf{r}-\mathbf{r}'|} &= \frac{1}{r}(1+\varepsilon)^{-\frac{1}{2}} = \frac{1}{r}\left(1-\frac{1}{2}\varepsilon+\frac{3}{8}\varepsilon^2-\frac{5}{16}\varepsilon^3+\dots\right) \\
&= \frac{1}{r}\left[1-\frac{1}{2}\left(\frac{r'}{r}\right)\left(\frac{r'}{r}-2\cos\theta'\right)+\frac{3}{8}\left(\frac{r'}{r}\right)^2\left(\frac{r'}{r}-2\cos\theta'\right)^2\right. \\
&\quad \left.-\frac{5}{16}\left(\frac{r'}{r}\right)^3\left(\frac{r'}{r}-2\cos\theta'\right)^3+\dots\right] \\
&= \frac{1}{r}\left[1-\frac{1}{2}\left(\frac{r'}{r}\right)^2+\left(\frac{r'}{r}\right)\cos\theta'+\frac{3}{8}\left(\frac{r'}{r}\right)^2\left(\left(\frac{r'}{r}\right)^2-4\left(\frac{r'}{r}\right)\cos\theta'+4\cos^2\theta'\right)\right. \\
&\quad \left.-\frac{5}{16}\left(\frac{r'}{r}\right)^3\left(\frac{r'}{r}-2\cos\theta'\right)\left(\left(\frac{r'}{r}\right)^2-4\left(\frac{r'}{r}\right)\cos\theta'+4\cos^2\theta'\right)+\dots\right] \\
&= \frac{1}{r}\left[1-\frac{1}{2}\left(\frac{r'}{r}\right)^2+\left(\frac{r'}{r}\right)\cos\theta'+\frac{3}{8}\left(\frac{r'}{r}\right)^4-\frac{3}{2}\left(\frac{r'}{r}\right)^3\cos\theta'+\frac{3}{2}\left(\frac{r'}{r}\right)^2\cos^2\theta'\right. \\
&\quad \left.-\frac{5}{16}\left(\frac{r'}{r}\right)^3\left\{\left(\frac{r'}{r}\right)^3-4\left(\frac{r'}{r}\right)^2\cos\theta'+4\left(\frac{r'}{r}\right)\cos^2\theta'-2\left(\frac{r'}{r}\right)\cos\theta'\right.\right. \\
&\quad \left.\left.+8\left(\frac{r'}{r}\right)\cos^2\theta'-8\cos^3\theta'\right\}+\dots\right] \\
&= \frac{1}{r}\left[1-\frac{1}{2}\left(\frac{r'}{r}\right)^2+\left(\frac{r'}{r}\right)\cos\theta'+\frac{3}{8}\left(\frac{r'}{r}\right)^4-\frac{3}{2}\left(\frac{r'}{r}\right)^3\cos\theta'+\frac{3}{2}\left(\frac{r'}{r}\right)^2\cos^2\theta'\right. \\
&\quad \left.-\frac{5}{16}\left(\frac{r'}{r}\right)^6+\frac{5}{4}\left(\frac{r'}{r}\right)^5\cos\theta'-\frac{5}{4}\left(\frac{r'}{r}\right)^4\cos^2\theta'+\frac{5}{8}\left(\frac{r'}{r}\right)^5\cos\theta'\right. \\
&\quad \left.-\frac{5}{2}\left(\frac{r'}{r}\right)^4\cos^2\theta'+\frac{5}{2}\left(\frac{r'}{r}\right)^3\cos^3\theta'+\dots\right] \\
&= \frac{1}{r}\left[1+\left(\frac{r'}{r}\right)(\cos\theta')+\left(\frac{r'}{r}\right)^2\left(-\frac{1}{2}+\frac{3}{2}\cos^2\theta'\right)\right. \\
&\quad \left.+\left(\frac{r'}{r}\right)^3\left(-\frac{3}{2}\cos\theta'+\frac{5}{2}\cos^3\theta'\right)+\dots\right] \\
&= \frac{1}{r}\left[1+\left(\frac{r'}{r}\right)(\cos\theta')+\left(\frac{r'}{r}\right)^2\left(\frac{3\cos^2\theta'-1}{2}\right)\right. \\
&\quad \left.+\left(\frac{r'}{r}\right)^3\left(\frac{5\cos^3\theta'-3\cos\theta'}{2}\right)+\dots\right] \tag{A.10}
\end{aligned}$$

In the last step of eq. (A.10), terms of $\left(\frac{r'}{r}\right)$ with like powers are collected and surprisingly, the coefficients are given by the Legendre polynomials. Rewriting eq. (A.10) in terms of Legendre polynomials:

$$\frac{1}{|\mathbf{r}-\mathbf{r}'|} = \frac{1}{r} \sum_{n=0}^{\infty} \left(\frac{r'}{r}\right)^n P_n(\cos\theta') \tag{A.11}$$

Substituting this back to eq. (A.8):

$$V(\mathbf{r}) = \frac{1}{4\pi\epsilon_0} \sum_{n=0}^{\infty} \frac{1}{r^{n+1}} \int (r')^n P_n(\cos \theta') \rho(\mathbf{r}') d\tau' \quad (\text{A.12})$$

The expression is exact in eq. (A.12), but this expansion is mainly used as an approximation techniques as the lowest non-zero terms in the expansion provide the approximate potential at large r and as greater precision is required, more terms from the expansion are included. The first term in the expansion is the monopole contribution (goes like $\frac{1}{r}$), second term is the dipole term (goes like $\frac{1}{r^2}$), the third term is the quadrupole term (goes like $\frac{1}{r^3}$) and so on. Thus, we see that the higher order terms contribute less to the potential at a given r .

The monopole and dipole terms

Expanding out eq. (A.12) for the first few terms:

$$\begin{aligned} V(\mathbf{r}) = & \frac{1}{4\pi\epsilon_0} \left[\frac{1}{r} \int \rho(\mathbf{r}') d\tau' + \frac{1}{r^2} \int r' \cos \theta' \rho(\mathbf{r}') d\tau' \right. \\ & \left. + \frac{1}{r^3} \int (r')^2 \left(\frac{3}{2} \cos^2 \theta - \frac{1}{2} \right) \rho(\mathbf{r}') d\tau' + \dots \right] \end{aligned} \quad (\text{A.13})$$

Let's examine the first two leading terms. The monopole term is given by:

$$\begin{aligned} V_{mon}(\mathbf{r}) &= \frac{1}{4\pi\epsilon_0} \frac{1}{r} \int \rho(\mathbf{r}') d\tau \\ &= \frac{1}{4\pi\epsilon_0} \frac{Q}{r} \end{aligned} \quad (\text{A.14})$$

where Q is the total charge of the configuration. The next term, dipole term is given by:

$$V_{dip}(\mathbf{r}) = \frac{1}{4\pi\epsilon_0} \frac{1}{r^2} \int r' \cos \theta' \rho(\mathbf{r}') d\tau' \quad (\text{A.15})$$

Notice that $r' \cos \theta' = \hat{\mathbf{r}} \cdot \mathbf{r}'$, then:

$$\begin{aligned} V_{dip}(\mathbf{r}) &= \frac{1}{4\pi\epsilon_0} \frac{1}{r^2} \hat{\mathbf{r}} \cdot \int \mathbf{r}' \rho(\mathbf{r}') d\tau' \\ &= \frac{1}{4\pi\epsilon_0} \frac{\mathbf{p} \cdot \hat{\mathbf{r}}}{r^2} \end{aligned} \quad (\text{A.16})$$

where $\mathbf{p} \equiv \int \mathbf{r}' \rho(\mathbf{r}') d\tau'$ is called the dipole moment of the distribution. For a collection of charges then the dipole moment is:

$$\mathbf{p} = \sum_{i=1}^N q_i \mathbf{r}'_i \quad (\text{A.17})$$

Recall the physical dipole example, the dipole moment using this formula is given by:

$$\begin{aligned} \mathbf{p} &= q\mathbf{r}'_+ - q\mathbf{r}'_- \\ &= q(\mathbf{r}'_+ - \mathbf{r}'_-) \\ &= q\mathbf{d} \end{aligned} \quad (\text{A.18})$$

Substituting this into eq. (A.16) then we recover eq. (A.7) as before.

In the case of a neutral atom, the total charge distribution is approximately zero, thus the monopole term will be approximately zero and the leading term in the multipole expansion is the dipole term. For example, when the wavelength of the electromagnetic radiation which induces, or is emitted during, transitions between different atomic energy levels is much larger than the typical size of a light atom, then keeping only the dipole term from the expansion is usually a good approximation to the solution. This approximation is known as the electric dipole approximation.

APPENDIX B

DERIVING THE EQUATIONS OF MOTION FOR THE WAVE FUNCTION

There are two ways to expand the wave function:

$$\Psi(r, t) = \sum_n C_n(t) u_n(r) e^{-i\omega_n t} \quad (\text{B.1})$$

$$\Psi(r, t) = \sum_n c_n(t) u_n(r) \quad (\text{B.2})$$

Consider a Hamiltonian:

$$H = H_0 + V \quad (\text{B.3})$$

where H_0 is the unperturbed Hamiltonian and V is the perturbation term that could be for example the interaction of a system with electromagnetic field.

The Schrodinger's equation is given by:

$$i\hbar \frac{\partial}{\partial t} \psi(\mathbf{r}, t) = H \psi(\mathbf{r}, t) \quad (\text{B.4})$$

Consider first the wave function expanded through eq. (B.1). Substituting eq. (B.1) and eq. (B.3) into eq. (B.4),

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \left[\sum_n C_n(t) u_n(\mathbf{r}) e^{-i\omega_n t} \right] &= [H_0 + V] \sum_n C_n(t) u_n(\mathbf{r}) e^{-i\omega_n t} \\ i\hbar \sum_n \left[-C_n(t) u_n(\mathbf{r}) i\omega_n e^{-i\omega_n t} + \dot{C}_n(t) u_n(\mathbf{r}) e^{-i\omega_n t} \right] &= \sum_n [\hbar\omega_n + V] C_n(t) u_n(\mathbf{r}) e^{-i\omega_n t} \\ \sum_n i\hbar \dot{C}_n(t) u_n(\mathbf{r}) e^{-i\omega_n t} &= V \sum_n C_n(t) u_n(\mathbf{r}) e^{-i\omega_n t} \end{aligned} \quad (\text{B.5})$$

Recall orthonormality condition:

$$\int u_n^*(\mathbf{r}) u_m(\mathbf{r}) d^3 r = \delta_{n,m} \quad (\text{B.6})$$

where $\delta_{n,m} = 1$ if $n = m$ and $\delta_{n,m} = 0$ if $n \neq m$.

Now, multiply both sides of eq. (B.5) from the left by u_m^* and integrate both sides over all space:

$$\begin{aligned} \int i\hbar \sum_n \dot{C}_n(t) u_m^*(\mathbf{r}) u_n(\mathbf{r}) e^{-i\omega_n t} d^3 r &= \int u_m^*(\mathbf{r}) V \sum_n C_n(t) u_n(\mathbf{r}) e^{-i\omega_n t} d^3 r \\ i\hbar \sum_n \dot{C}_n(t) e^{-i\omega_n t} \delta_{n,m} &= \sum_n \int d^3 r u_m^*(\mathbf{r}) V u_n(\mathbf{r}) e^{-i\omega_n t} C_n(t) \\ i\hbar \dot{C}_m(t) e^{-i\omega_m t} &= \sum_n \langle m|V|n\rangle e^{-i\omega_n t} C_n(t) \end{aligned} \quad (\text{B.7})$$

where $\langle m|V|n\rangle = \int d^3 r u_m^*(\mathbf{r}) V u_n(\mathbf{r})$. Finally multiply both sides of eq. (B.7) by $e^{i\omega_m t}$ and rearranging:

$$\dot{C}_m(t) = -\frac{i}{\hbar} \sum_n \langle m|V|n\rangle e^{i\omega_{mn} t} C_n(t) \quad (\text{B.8})$$

where $\omega_{mn} = \omega_m - \omega_n$.

Applying exactly the same procedure to eq. (B.2):

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} [\sum_n c_n(t) u_n(\mathbf{r})] &= [H_0 + V] \sum_n c_n(t) u_n(\mathbf{r}) \\ i\hbar \sum_n \dot{c}_n(t) u_n(\mathbf{r}) &= \sum_n (\hbar\omega_n + V) c_n(t) u_n(\mathbf{r}) \\ u_m^* \int i\hbar \sum_n \dot{c}_n(t) u_n(\mathbf{r}) d^3 r &= u_m^* \sum_n \int (\hbar\omega_n + V) c_n(t) u_n(\mathbf{r}) d^3 r \\ i\hbar \dot{c}_m(t) &= \sum_n \hbar\omega_n \int d^3 r u_m^* u_n(\mathbf{r}) c_n(t) + \sum_n \int d^3 r u_m^* V u_n(\mathbf{r}) c_n(t) \\ c_m(t) &= -\frac{i}{\hbar} [\hbar\omega_m c_m(t) + \sum_n \langle m|V|n\rangle c_n(t)] \\ \dot{c}_m(t) &= -i\omega_m c_m(t) - \frac{i}{\hbar} \sum_n \langle m|V|n\rangle c_n(t) \end{aligned} \quad (\text{B.9})$$

Eq. (B.8) and eq. (B.9) derived from eq. (B.1) and eq. (B.2) are equivalent to the original Schrodinger's equation. In deriving eq. (B.8) the time dependent part of the unperturbed Hamiltonian cancels out, thus, eq. (B.8) only depends on the perturbed part of the Hamiltonian (interaction term). Thus, eq. (B.8) is typically called the interaction

representation. On the other hand, eq. (B.9) depends on the total Hamiltonian (the unperturbed Hamiltonian does not cancel out in the derivation). Eq. (B.8) and eq. (B.9) are no longer concerned with the position of the eigenfunctions, but rather they describe the time evolution of the probability amplitudes of the total energy eigenfunctions. They are useful for example in describing a system of an atom absorbing energy from light field. Eq. (B.8) and eq. (B.9) gives a complete description of the development of such a system.

These two equations can be written in matrix notation. This description is especially useful for example in a multi-level atom system, the individual states and their interference terms are all described by their respective matrix elements.

Using eq. (B.9) and rewriting:

$$\begin{aligned}
 \dot{c}_m(t) &= -i\omega_m c_m(t) - \frac{i}{\hbar} \sum_n \langle m|V|n\rangle c_n(t) \\
 i\hbar \dot{c}_m(t) &= \hbar\omega_m c_m(t) + \sum_n \langle m|V|n\rangle c_n(t) \\
 i\hbar \dot{\mathbf{c}}_m(t) &= \mathbf{E}_m c_m(t) + \mathbf{V}(t) \mathbf{c}_n(t)
 \end{aligned} \tag{B.10}$$

The V on the 2nd term on the RHS of eq. (B.10) has matrix elements V_{mn} and dimension $m \times m$ (since the dimension of the RHS has to be the same as the LHS). The first term in eq. (B.10) is a $m \times m$ matrix that has only diagonal terms that are non-zero since in deriving eq. (B.9), the orthogonality condition was applied and only terms of $n = m$ are picked out by the orthogonality condition. Thus, eq. (B.10) in matrix form:

$$i\hbar \dot{\mathbf{c}}_m(t) = \begin{pmatrix} E_1 & V_{12} & V_{13} & \dots & V_{1M} \\ V_{21} & E_2 & & & \dots \\ V_{31} & & \dots & & \\ \dots & & & & \\ V_{M1} & & & & E_M \end{pmatrix} \begin{pmatrix} c_1 \\ c_2 \\ \dots \\ c_M \end{pmatrix} \tag{B.11}$$

Notice that there is no summation term in eq. (B.11). This is because the summation is implied in the multiplication of the matrix and the row vector.

Similarly, using the equation of motion in the interaction picture, eq. (B.8) and rewriting:

$$\begin{aligned}\dot{C}_m(t) &= -\frac{i}{\hbar} \sum_n \langle m|V|n\rangle \mathbf{e}^{i\omega_{mn}t} C_n(t) \\ i\hbar \dot{C}_m(t) &= \sum_n \langle m|V|n\rangle \mathbf{e}^{i\omega_{mn}t} C_n(t)\end{aligned}\tag{B.12}$$

In matrix form, eq. (B.12) becomes:

$$i\hbar \dot{\mathbf{C}}_m(t) = \begin{pmatrix} 0 & V_{12}e^{i\omega_{12}t} & V_{13}e^{i\omega_{13}t} & \dots & V_{1M}e^{i\omega_{1M}t} \\ V_{21}e^{i\omega_{21}t} & 0 & & & \dots \\ V_{31}e^{i\omega_{31}t} & & \dots & & \\ \dots & & & & \\ V_{M1}e^{i\omega_{M1}t} & & & & 0 \end{pmatrix} \begin{pmatrix} c_1 \\ c_2 \\ \dots \\ c_M \end{pmatrix}\tag{B.13}$$

Notice that the diagonal terms are all zero. This is because the unperturbed Hamiltonian part was canceled out in the derivation of eq. (B.8).

REFERENCES

- Alnis, J., Blushs, K., Auzinsh, M., Kennedy, S., Shafer-Ray, N., & Abraham, E. R. I. (2003). The hanle effect and level crossing spectroscopy in rb vapour under strong laser excitation. *Journal of Physics B: Atomic, Molecular and Optical Physics*, 36(6), 1161.
- Fleischhauer, M., & Lukin, M. D. (2000, May). Dark-state polaritons in electromagnetically induced transparency. *Phys. Rev. Lett.*, 84, 5094–5097.
- Folk, J. A., Marcus, C. M., & Harris, J. S. (2001, Oct). Decoherence in nearly isolated quantum dots. *Phys. Rev. Lett.*, 87, 206802.
- Griffiths, D. J. (1999). *Introduction to electrodynamics*. Prentice-Hall.
- Hanle, W. (1924). Uber magnetische beeinflussung der polarisation der resonanzfluoreszenz. *Zeitschrift fr Physik*, 30, 93-105.
- Hatef, A., & Singh, M. R. (2010, Jun). Plasmonic effect on quantum coherence and interference in metallic photonic crystals doped with quantum dots. *Phys. Rev. A*, 81, 063816.
- Hau, L. V., Harris, S. E., Dutton, Z., & Behroozi, C. H. (1999, February). Light speed reduction to 17 metres per second in an ultracold atomic gas. *Nature*, 397(4), 594-598.
- Jackson, J. D. (1998). *Classical electrodynamics*. John Wiley and Sons, Inc.
- Marlan O. Scully, M. S. Z. (1997). *Quantum optics*. Cambridge University Press.
- Mikhailov, E. E. (2003). *Nonlinear Properties Of Dense Coherent Media*. Unpublished doctoral dissertation, Texas AM University, United States of America.
- Ooi, C. H. R., & Tan, K. S. (2013). Controlling double quantum coherence and electromagnetic induced transparency with plasmonic metallic nanoparticle. *Plasmonics*, 8(2), 891-898.
- Paul R. Berman, V. S. M. (2011). *Principle of laser spectroscopy and quantum optics*. Princeton University Press.
- Sadeghi, S. M., Deng, L., Li, X., & Huang, W.-P. (2009). Plasmonic (thermal) electromagnetically induced transparency in metallic nanoparticlequantum dot hybrid systems. *Nanotechnology*, 20(36), 365401.
- Savasta, S., Saija, R., Ridolfo, A., Di Stefano, O., Denti, P., & Borghese, F. (2010). Nanopolaritons: Vacuum rabi splitting with a single quantum dot in the center of a dimer nanoantenna. *ACS Nano*, 4(11), 6369-6376.
- Shimizu, K. T., Woo, W. K., Fisher, B. R., Eisler, H. J., & Bawendi, M. G. (2002, Aug). Surface-enhanced emission from single semiconductor nanocrystals. *Phys. Rev. Lett.*, 89, 117401.
- Whitley, R. M., & Stroud, C. R. (1976, October). Double optical resonance. *Physical Review A*, 14(4), 1498–1513.