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Optical processes of a single-atom model in the phonon field†

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Abstract. The optical processes of a single-atom model in the phonon field have been studied. To deal with the quantum electrodynamics phenomena in condensed matter microcavities, the influence of electron–phonon interactions has to be taken into account. As a clear picture, the effect of the lattice's oscillation on the optical transition in condensed matter microcavities is reduced to a model of a single atom in a phonon field. We deduce that in the case with a phonon participating, the Bloch equation is still satisfied. In the process of spontaneous emission for an electron interacting with a phonon, we derive the Rabi oscillation for the simple atom in the phonon field.

1. Introduction

Along with the development of optical communication and the microlaser, the atom–vacuum interaction in microcavities with dimensions of the order of a micron has become an attractive new topic called cavity quantum electrodynamics (cavity QED) [1–3]. Interactions between atoms and the vacuum in microcavities produce many interesting phenomena; for example, the spontaneous emission rate of atoms can be enhanced or forbidden in microcavities through changes of the dimension of the cavities [4], an atom can exchange energy periodically with a cavity, instead of radiatively decaying to a lower energy state, etc [5]. Recently, developments in molecular beam epitaxy and other growth techniques have made it possible to tailor quantum confined structures, such as quantum wells, superlattices, quantum lines, and quantum dots. Therefore, the microfabrication techniques make condensed matter microcavities, such as polymer spheres, or semiconductor Fabry–Perot microcavities, possible. Naturally, the investigation of the cavity QED effect in condensed matter microcavities is raised. When lightwaves propagate in a condensed matter microcavity, the electronic state of the atoms is changed, accompanied by radiation of the electromagnetic field which reacts back with the electron. The light and the excited electrons change the lattice vibrations, and the changed periodic potential of the lattice reacts with the electrons; in general, the interaction between the electrons and the lattice can be described by the interaction between the electrons and phonons. When the wavelength of the light is in the range of infra-red or far infra-red waves, this interaction has to be taken into account. If we only consider the linear interaction, there are at least three terms we have to deal with: the interaction between photon and electron, the interaction between electron and phonon, and that between photon and phonon.

In this paper, we report a single-atom model in which the effect of the lattice oscillation on the atom is reduced to a phonon field. For the processes involved, interaction between

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the radiation and condensed matter, we thus consider only a single atom located in a phonon field, instead of dealing with a complicated problem such as an atom in a periodic potential. For simplicity, this atom has only one electron and two non-degenerate energy levels; the electron can transit between the ground state and the excited state. In section 2, we will give the Hamiltonian for this system. In section 3, we show that the Bloch equation is still satisfied for this system. In section 4, we discuss the Rabi oscillation and Rabi frequency for this system.

2. The Hamiltonian of the system

In this section we present and discuss the assumptions constituting the theoretical model. The total Hamiltonian of the system for an atom in a light field and a phonon field is

$$H = H_e + H_{photon} + H_{phonon} + H_{I1} + H_{I2} + H_{I3} \tag{1}$$

where H_e is the electronic Hamiltonian, H_{photon} is the photon Hamiltonian, and H_{phonon} is the phonon Hamiltonian. H_{I1}, H_{I2}, and H_{I3} are the Hamiltonians for the interactions between electron and photon, electron and phonon, and photon and phonon, respectively. The Hamiltonian for this system, as written above, is based on the adiabatic approximation (Born–Oppenheimer method). Because electrons and ions have very different masses, the ions can only respond slowly to a change in the electron configuration, while the electrons respond adiabatically to a change in the position of the ions. Then we can divide the total Hamiltonian into those for the movement of the electrons within a stationary lattice and those for the movement of the ions in a uniform space containing electrons, i.e. the term for the phonons. Next we will deduce their expressions in the representation of the interaction.

(i) The electron Hamiltonian.

The energy of the excited state $|i\rangle$ is indicated by E_i ($i = 1, 2$ for our mode, i.e. the ground state and the excited state). It is convenient to work with the Hamiltonian expressed in the representation of second quantization. Let $\phi_j(r)$ be the Schrödinger eigenfunction for the electrons, then any wavefunctions can be expressed using this set of basic functions

$$\Psi(r,t) = \sum_j b_j \phi_j(r) \qquad \Psi^*(r,t) = \sum_j b_j^\dagger \phi_j^*(r)\,.$$

Thus the creation and annihilation operators of electrons in the states $|j\rangle$ are denoted by $b_j^\dagger$ and b_j, respectively. The Hamiltonian of the electrons is then

$$H_e = \sum_j \hbar\omega_i (b_j^\dagger b_j + \tfrac{1}{2})\,. \tag{2}$$

The creation and annihilation operators for electrons are Fermi operators and they have the following properties:

$$b_j|0\rangle = 0 \tag{3}$$

where $|0\rangle$ is the vacuum state. The electron in the jth state is denoted by

$$b_j^\dagger|0\rangle\,. \tag{4}$$

By the Pauli exclusion principle, there are no two electrons in one state, therefore

$$b_j^\dagger b_j^\dagger|\phi\rangle = 0\,. \tag{5}$$

The commutation relations of the Fermi operators are

$$[b_j^\dagger, b_k]_+ = \delta_{jk} \qquad [b_j^\dagger, b_k^\dagger]_+ = 0 \qquad [b_j, b_k]_+ = 0 \tag{6}$$

where $[a, b]_+ = ab + ba$.

(ii) The phonon Hamiltonian.

Assuming that the crystal is non-conducting and the lattice dynamics are harmonic, the lattice thermal oscillations can be considered as a summation of a set of normal modes, the creation and annihilation operators of phonons are denoted by $c_\Lambda^\dagger(q)$ and $c_\Lambda(q)$, where Λ is the index of the mode, whose frequency is ω_Λ, and q is the wavevector of phonon in reciprocal space. These operators lead to a phonon Hamiltonian

$$H_{\text{phonon}} = \sum_\Lambda \hbar\omega_\Lambda (c_\Lambda^\dagger c_\Lambda + \tfrac{1}{2}) . \tag{7}$$

The phonon field described by (7), which is a collective excitation, consists of non-interacting phonons. Each state in the lattice vibration spectrum can be occupied by any number of phonons, since phonons are bosons.

(iii) The photon Hamiltonian.

The quantization of the electromagnetic field leads to the introduction of photons. We denote the creation and annihilation operators for the photons by $a_\lambda^\dagger$ and a_λ, where λ is the index of the field mode. The Hamiltonian for the photons is

$$H_{\text{photon}} = \sum_\lambda \hbar\omega_\lambda (a_\lambda^\dagger a_\lambda + \tfrac{1}{2}) . \tag{8}$$

Both photon and phonon follow Bose statistics. The creation and annihilation operators for Bosons have the following properties:

$$a|n\rangle = 0 \qquad a|n\rangle = \sqrt{n}|n-1\rangle \qquad a^\dagger|n\rangle = \sqrt{n+1}|n+1\rangle \qquad a^\dagger a|n\rangle = n|n\rangle \tag{9}$$

where $a_\lambda^\dagger$ and a_λ fulfill the commutation relation

$$[a_\lambda, a_{\lambda'}^\dagger] = \delta_{\lambda\lambda'} \qquad [a_\lambda, a_\lambda] = [a_\lambda^\dagger, a_\lambda^\dagger] = 0 . \tag{10}$$

(iv) The photon–electron interaction.

Using the electric dipole momentum approximation, and neglecting the higher terms, we have the Hamiltonian of the interaction between electron and electromagnetic field [6]:

$$H_{I1} = \hbar \sum_{jk\lambda} b_j^\dagger b_k g_{\lambda,jk} (a_\lambda^\dagger + a_\lambda) \tag{11}$$

where $g_{\lambda,jk}$, the coupling coefficient, is given, apart from unimportant factors, by

$$g_{\lambda,jk} = \int \phi_j^*(r) |u_\lambda p| \phi_k(r) \mathrm{d}r \tag{12}$$

where p is the momentum and u is the normal coordinate of the system, and $\phi_j(r)$, $\phi_k(r)$ are the eigenfuctions of the Schrödinger equation for electrons in the states j and k, respectively. Here we assume the electron field and the electromagnetic field are independent of each other; their operators, therefore, commute with each other, and we have

$$[a_\lambda^\dagger, b_j^\dagger] = 0 \qquad [a_\lambda, b_j] = 0 \qquad [a_\lambda^\dagger, b_j] = 0 \qquad [a_\lambda, b_j^\dagger] = 0 . \tag{13}$$

(v) The electron–phonon interaction.

Electrons within a crystal give rise to a certain charge distribution which produces an electric field that influences the lattice vibrations. Since the field of the electron is a static field, it only interacts with the longitudinal acoustic phonons [7]. The basic electron–phonon interaction process is absorption or emission of a phonon with a simultaneous change of the electron state. The interaction Hamiltonian is [7]

$$H_{I2} = \sum_{\lambda\lambda'} \gamma_{\lambda\lambda'\Lambda} b_\lambda^\dagger b_{\lambda'} (c_\Lambda + c_\Lambda^\dagger) \tag{14}$$

where the coupling coefficient $\gamma_{\lambda\lambda'\Lambda}$ is given by

$$\gamma_{\lambda\lambda'\Lambda} \equiv \int \mathrm{d}r\, \phi_\lambda^*(r) \gamma_\Lambda \phi_{\lambda'}(r) \tag{15}$$

and γ_Λ, given by $H_{I2} = -\sum_\Lambda \gamma_\Lambda(r)(c_\Lambda + c_\Lambda^\dagger)$, is the Taylor expantion coefficient of H_{I2} related to the electron, and $\phi_\lambda(r)$ is the eigenfunction for the electron.

(vi) The photon–phonon interaction.

Since the polarized field of a transverse optical phonon is a curled field, an electromagnetic field is accompanied by a transverse optical phonon. Thus the photon will interact with the transverse optical phonon. There are many different processes; for instance, first-order processes involve absorption of only one photon and one phonon of identical energy and wavevector generation. In a secondary process a photon decays into two or more phonons. In the process of light scattering, a photon transforms another photon of different energy with absorption or emission of one or more phonons. If the phonon involved in such processes belongs to the optical branch then we have Raman scattering, if they belong to the acoustic branch we have Brilliouin scattering. In our simple model, we only consider the first-order process. Classically, the interaction energy is due to a polarization vibrator in the electric field, therefore the interaction energy is the negative of the product of the polarization vector and the electric field. Then the Hamiltonian of this interaction is [8]

$$H_{I3} = -\boldsymbol{p}\cdot\boldsymbol{E} \sim \sum_{qn} (c_{-q}^\dagger + c_q) \exp[i\boldsymbol{q}\cdot\boldsymbol{R}_n + \boldsymbol{k}\cdot\boldsymbol{r})]\,. \tag{16}$$

Here we assume that the interaction between photon and phonon is much smaller than that between electron and phonons, therefore we neglect the term expressed in (16) when we go further.

3. Quantum Bloch equation

The atom in which the electrons are in different electronic states can be regarded as if the atom is in different atomic states. For convenience, we introduce the atomic operators to indicate the change of atomic states. Respectively, we use the upper and lower operators $S^{(+)}$ and $S^{(-)}$, which denote the transition of the electron excited from the ground state to the excited state and from the excited state to the ground state:

$$S^{(+)} = |2\rangle\langle 1| \qquad S^{(-)} = |1\rangle\langle 2| \tag{17}$$

where $|1\rangle$ and $|2\rangle$ show the ground state and excited state, respectively. For convenience, we take the zero point of energy as the mid-point between two levels, i.e. $E_2 = -E_1 = \frac{1}{2}\hbar\omega_0$. The operator S_z can be used to describe the Hamiltonian of the two-level free atomic state,

$$H_{\text{atom}} = \hbar\omega_0(b^\dagger b) = \hbar\omega_0 S_z \tag{18}$$

where ω_0 is given by $\hbar\omega_0 = E_2 - E_1$, $S_z = \frac{1}{2}\sigma_z$ and σ_z is the Pauli operartor. The atomic operators fulfill the commutation relations

$$[S^{(+)}, S^{(-)}] = 2S_z \qquad [S^{(\pm)}, S_z] = \mp S^{(\pm)}. \tag{19}$$

Using the atomic operators, we can rewrite the Hamiltonian of a two-level atom and multimode photon and phonon field system as

$$H = \hbar\omega_0 S_z + \sum_\lambda \hbar\omega_\lambda a_\lambda^\dagger a_\lambda + \sum_\Lambda \hbar\omega_\Lambda c_\Lambda^\dagger c_\Lambda + \hbar(gS^{(-)} + g^* S^{(+)})A + \sum_\Lambda \hbar\gamma_\Lambda S_z C \tag{20}$$

where $A = \sum_\lambda (a_\lambda^\dagger + a_\lambda)$, $C = \sum_\Lambda (c_\Lambda^\dagger + c_\Lambda)$, and γ_Λ is a coefficient. In the Heisenberg representation, the equation of motion for an operator O is

$$i\hbar \frac{dO}{dt} = [O, H]. \tag{21}$$

Then we can induce the following equations with the commutation relation of (18):

$$\begin{aligned} \frac{dS_z}{dt} &= i[gS^{(-)} - g^* S^{(+)}]A \\ \frac{dS^{(+)}}{dt} &= i[S^{(+)}\omega_0 + S^{(+)}C - 2gS_z A] \\ \frac{dS^{(-)}}{dt} &= -i[S^{(-)}\omega_0 + S^{(-)}C - 2g^* S_z A]. \end{aligned} \tag{22}$$

We introduce the following variables:

$$\begin{aligned} R_1 &= -(g^* S^{(+)} + gS^{(-)}) \\ R_2 &= i(g^* S^{(+)} - gS^{(-)}) \\ R_3 &= 2|g|S_z. \end{aligned} \tag{23}$$

From (22), we can get

$$\begin{aligned} \dot{R}_1 &= -i(\omega_0 + C)R_2 \\ \dot{R}_2 &= -(\omega_0 + C)R_1 + KAR_3 \\ \dot{R}_3 &= -KAR_2 \end{aligned} \tag{24}$$

where $K = 2|g|$. We next define the nutation vector, $\boldsymbol{R}$, and rotation momentum vector, Ω, as

$$\boldsymbol{R} = R_1\hat{x} + R_2\hat{y} + R_3\hat{z} \tag{25}$$

$$\Omega = -KA(t)\hat{x} + (\omega_0 + C)\hat{z}. \tag{26}$$

Thus equation (24) can be written as the Bloch equation:

$$\frac{dR(t)}{dt} = \Omega(t) \times \boldsymbol{R}(t). \tag{27}$$

The vector $\boldsymbol{R}$ describes the atomic states, which rotate around the vector Ω, which depends on the photon and phonon field. Thus, considering the electron–phonon interaction, the Bloch equation is still satisfied, except the components of $\boldsymbol{R}$ and Ω are changed.

4. Spontanous emission

Now we consider the system of a two-level atom in the single-mode photon field and the single-mode phonon field. By the rotation-wave approximation, the system Hamiltonian can be written as

$$H = \hbar\omega_0 S_z + \hbar\omega_1 a^\dagger a + \hbar\omega_2 c^\dagger c + \hbar(g a^\dagger S^{(-)} + g^* S^{(+)} a) + \hbar\gamma S_z(c^\dagger + c) \tag{28}$$

where ω_0, ω_1, and ω_2 are the frequencies of the electron, photon and phonon, respectively. As mentioned before, electrons only couple with the longitudinal acoustic (LA) phonons, which are elastic waves in the crystal. The energy of the LA phonons is much less than the excitation energy of the electrons, therefore they have no contribution to the electronic excitation. Hence, in this system, the excitation number operators $N_1 = S_z + a^\dagger a + c^\dagger c$, as well as $N_2 = S_z + a^\dagger a - c^\dagger c$, are conserved and they commute with the system Hamiltonian, which can be rewritten as

$$H = \tfrac{1}{4}\hbar\overline{\omega}_1 N_1 + \tfrac{1}{4}\hbar\overline{\omega}_2 N_2 + \tfrac{1}{4}\hbar\overline{\omega}_3 N_3 + \tfrac{1}{4}\hbar\overline{\omega}_4 N_4 + \hbar(g a^\dagger S^{(-)} + g^* S^{(+)} a) + \hbar\gamma S_z(c^\dagger + c) \tag{29}$$

where

$$\begin{aligned} N_1 &\equiv (S_z + a^\dagger a + c^\dagger c) & N_2 &\equiv (S_z + a^\dagger a - c^\dagger c) \\ N_3 &\equiv (S_z - a^\dagger a + c^\dagger c) & N_4 &\equiv (S_z - a^\dagger a - c^\dagger c) \end{aligned} \tag{30}$$

and

$$\begin{aligned} \overline{\omega}_1 &\equiv \omega_1 + \omega_2 + \omega_3 & \overline{\omega}_2 &\equiv \omega_1 + \omega_2 - \omega_3 \\ \overline{\omega}_3 &\equiv \omega_1 - \omega_2 + \omega_3 & \overline{\omega}_4 &\equiv \omega_1 - \omega_2 - \omega_3 . \end{aligned} \tag{31}$$

In the Heisenbeger representation, the equation of motion of the atomic operator is

$$\dot{S}_z = -\frac{\mathrm{i}}{\hbar}[S_z, H] = \mathrm{i}(g a^\dagger S^{(-)} - g^* S^{(+)} a) \tag{32}$$

$$\begin{aligned} \ddot{S}_z = -\frac{\mathrm{i}}{\hbar}[\dot{S}_z, H] &= \frac{1}{\hbar} g[a^\dagger S^{(-)}, H] - \frac{1}{\hbar} g^*[S^{(+)} a, H] \\ &= \tfrac{1}{2}[\overline{\omega}_3 + \overline{\omega}_4 + 2\gamma(c^\dagger + c)](g a^\dagger S^{(-)} + g^* S^{(+)} a) - 4|g|^2(\tfrac{1}{2} + a^\dagger a + S_z) S_z . \end{aligned} \tag{33}$$

To evaluate these formule, we used the relations, $S^{(+)} S^{(-)} = S_z + \frac{1}{2}$ and $S_z^2 = \frac{1}{4}$, and the commutation relations in (19). Letting $\omega^R = H/\hbar$, we can simplify equation (33) to

$$\left(\frac{\mathrm{d}^2}{\mathrm{d}t^2} + \Omega^2\right) S_z = X\left(\omega^R - \tfrac{1}{4}[N_1(\overline{\omega}_1 - \overline{\omega}_4) + N_2(\overline{\omega}_2 - \overline{\omega}_3)]\right) \tag{34}$$

where the quantity X is given by

$$X = \tfrac{1}{2}[\overline{\omega}_3 + \overline{\omega}_4 + 2\gamma(c^\dagger + c)] \tag{35}$$

and the operator Ω is given by

$$\Omega^2 = X^2 + 4|g|^2(\tfrac{1}{2} + a^\dagger a + S_z) . \tag{36}$$

It is clear that Ω is the Rabi frequency, at which the atom reverses between the ground state and the excitation state.

Since the right-hand side of (34) is the conserved quantity, it can be solved. Let $|n, m, l\rangle = |n\rangle|m\rangle|l\rangle$ be the eigenstate of $a^\dagger a$, S_z and $c^\dagger c$, i.e.

$$\begin{aligned} &a^\dagger(0)a(0)|n, m, l\rangle = n|n, m, l\rangle \qquad n = 0, 1, \ldots \\ &S_z(0)|n, m, l\rangle = m|n, m, l\rangle \qquad m = \tfrac{1}{2} \quad \text{or} \quad -\tfrac{1}{2} \\ &c^\dagger c|n, m, l\rangle = l|n, m, l\rangle \qquad l = 0, 1, \ldots . \end{aligned} \tag{37}$$

It is easy to obtain $\langle\omega^R\rangle = m\omega_0 + n\omega_1 + l\omega_2$ and $\langle N_1\rangle = n + m + l$; the solution of (34) is then

$$\langle S_z(t, X)\rangle = \langle S_z(0, X)\rangle\left[1 - \frac{8|g|^2(n + m + \frac{1}{2})}{X^2 + 4|g|^2(n + m + \frac{1}{2})}\sin^2\tfrac{1}{2}\Omega t\right]. \tag{38}$$

For the spontanous emission in the vaccum electromagnetic field, $n = 0$, $m = \frac{1}{2}$ and we have

$$\langle S_z(t, X)\rangle = \frac{1}{2}\left[1 - \frac{2q^2}{X^2 + q^2}\sin^2\frac{1}{2}\sqrt{X^2 + q^2}t\right] \tag{39}$$

where $q = 4|g|^2$.

From equations (38) and (39), we can draw the following important conclusions.

(i) The Bloch equation is satisfied even in the case of the phonon participating, but, due to the electron–phonon interaction, there is an extra term in the parameter of the Bloch equation.

(ii) When the atom interacts with the photon and phonon fields, it will be excited from the ground state to the state which is the superposition of the ground state and excited state, and then it returns to the ground state.

(iii) From equation (35), the contribution of phonons to the excitation of the electrons is an addition to the Rabi resonance frequency.

(iv) From (14) and (15), the coupling coefficients g and γ are related to r, the position of the electrons; therefore, the Rabi oscillation frequency depends on the position vector of the electrons.

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