

Effect of the counterrotating terms on polarizability in atom-field interactions

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The effect of the counterrotating terms on the linear polarizability is investigated, which is responsible for the validity of the optical theorem in all frequency regions. A unitary transformation method [H. Zheng, S. -Y. Zhu, and M.S. Zubairy, *Rev. Lett.* **101**, 200404 (2008)] is adopted to overcome the difficulty brought in by the counterrotating terms, which yields a rotating-wave-approximation-like Hamiltonian with modified coupling constant due to the counterrotating terms. A simple expression for the polarizability is obtained, which is a sum of resonant (minus sign) and antiresonant (plus sign) parts, and from which the role of the counterrotating terms and quantum interference between the counterrotating terms and rotating terms at far off-resonance are discussed.

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I. INTRODUCTION

In the study of the atom-field interaction, the absorption and scattering of the light are two aspects of the same physical process, which are expressed by the imaginary part of the polarizability $\text{Im } \alpha(\omega)$ and scattering cross section $\sigma_{cs}(\omega)$, respectively. Energy conservation requires the rate of removal of energy from the incident light due to absorption to be equal to the rate of energy increase in the scattered field [1,2],

$$\frac{\omega}{\epsilon_0 c} \text{Im } \alpha(\omega) = \frac{\omega^4}{2\pi\epsilon_0^2 c^4} |\alpha(\omega)|^2 = \sigma_{cs}(\omega). \quad (1)$$

Equation (1) describes the relation between the polarizability and the scattering cross section, which is the well-known optical theorem. For a two-level atom with transition frequency ω_0 , the general expression of the complex polarizability according to the Kramers-Heisenberg formula [3,4] is

$$\alpha(\omega) = \frac{d^2}{3\hbar} \left\{ \frac{1}{\omega_0 - \omega - i\gamma_-(\omega)} + \frac{1}{\omega_0 + \omega + i\eta\gamma_+(\omega)} \right\}, \quad (2)$$

where $\gamma_-(\omega)$ [$\gamma_+(\omega)$] is the resonant (antiresonant) damping rate. The η value depends on approximations used in the calculation, $\eta=0$ [3,5] or $\eta=+1$ [3]. Besides, $\eta=-1$ is also suggested in some studies (see the discussion in [3]). In Ref. [5], the rotating-wave-approximation (RWA) is adopted for the atom-vacuum Hamiltonian. This Hamiltonian is first diagonalized and then the diagonalized Hamiltonian interacts with the external light. This yields an expression with

$$\gamma_{\pm}(\omega) = \frac{d^2 \omega^3}{6\pi\epsilon_0 \hbar c^3} \theta(\mp \omega),$$

where $\theta(\omega)$ is the unit step function and ω is the external field frequency. In this method, the imaginary part of the polarizability satisfies the frequency-dependent property required by the optical theorem, but still does not satisfy the optical theorem, particularly in the nonresonant case. In Ref. [3], the polarizability is calculated in the Heisenberg picture

under the RWA, and the same result as in Ref. [5] is obtained. The authors also generalize the result beyond RWA and $\eta=+1$ is suggested. However the result does not satisfy the optical theorem. In Ref. [1], the polarizability is recalculated without the RWA by using the ground state of the whole atom-vacuum system with a Green's function method and a result satisfying the optical theorem is obtained. This method has been applied recently to the three-level atom recently [4]. In Ref. [2], the polarizability without RWA is calculated in both Schrödinger and Heisenberg pictures, which, instead of the Markovian approximation as in Ref. [3], uses the formal solution of the Heisenberg equation of motion for the Pauli matrix $\sigma_z(t)$ and also goes “beyond the standard Weisskopf-Wigner approximation” [3], which means without RWA and Markov approximation. The obtained result satisfies the optical theorem. However, the expression for the polarizability is very complicated compared to Eq. (2), and could not be simplified to analyze the role of the counterrotating terms on the polarizability. In a recent study [6], the polarizability is calculated by using Feynman diagrams and propagators, which can be reduced to the form in [2] (see the discussions in Ref. [4]). In addition, the authors of Ref. [6] also argued that $\eta=+1$ should be applied for the linear polarizability, while $\eta=-1$ should be applied in the scattering situation. In Refs. [1,2,6], the polarizability is not expressed in the form of Eq. (2) and therefore, it is hard to see which prescription the results support.

We can conclude from the previous studies that one must go beyond the RWA and base the calculations on the ground state of the whole atom-vacuum system in order to obtain the right expression for polarizability which satisfies the optical theorem. However all the foregoing methods are too cumbersome to study the effects of the counterrotating terms on the polarizability. These methods can thus be hardly extended to the true multilevel atomic system.

In this paper, we use a method in the Heisenberg picture to obtain a simple equation for the polarizability without the RWA. The result satisfies the optical theorem. Our calculations reveal the role of the counterrotating terms and the physics behind them. We first make a unitary transformation of the total Hamiltonian including the counterrotating terms

to obtain an effective Hamiltonian where the interaction part has the same form as under RWA [7]. The polarizability can be obtained in a straightforward manner starting with the transformed Hamiltonian. The resultant polarizability satisfies the optical theorem up to the second order of the atom-vacuum coupling constant.

The paper is organized as following. In Sec. II, we make a unitary transformation of the total Hamiltonian and go beyond RWA in the Heisenberg picture; in Sec. III, we discuss the physical effect of the counterrotating terms, and in Sec. IV, we have a brief discussion about the frequency shift; Finally, in Sec. V, we summarize the main point of this paper.

II. POLARIZABILITY OF A TWO-LEVEL ATOM WITHOUT RWA

The Hamiltonian for the system of a two-level atom with transition frequency ω_0 coupled with vacuum field is as following ($\hbar=1$):

$$H = \frac{1}{2}\omega_0\sigma_z + \sum_{\mathbf{k}} \omega_{\mathbf{k}}a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \sum_{\mathbf{k}} g_{\mathbf{k}}(a_{\mathbf{k}}^\dagger + a_{\mathbf{k}})(\sigma^+ + \sigma), \quad (3)$$

where σ_z is the z component of the Pauli matrix, and σ and σ^+ are the atomic raising and lowering operator, respectively. $a_{\mathbf{k}}$ and $a_{\mathbf{k}}^\dagger$ are the annihilation and creation operators for the \mathbf{k} th vacuum mode. $g_{\mathbf{k}} = \mathbf{d} \cdot \hat{\mathbf{e}}_{\mathbf{k}}(\omega_{\mathbf{k}}/2\epsilon_0 V)^{1/2}$ is the coupling strength between the \mathbf{k} th vacuum mode with unit polarization vector $\hat{\mathbf{e}}_{\mathbf{k}}$ and atom with dipole moment \mathbf{d} , and the summation over \mathbf{k} includes two polarizations. V is the quantization volume. After making the unitary transformation of the Hamiltonian $H^S = e^{iS} H e^{-iS}$, with

$$S = \sum_{\mathbf{k}} \frac{g_{\mathbf{k}} \xi_{\mathbf{k}}}{i\omega_{\mathbf{k}}} (a_{\mathbf{k}}^\dagger - a_{\mathbf{k}})(\sigma^+ + \sigma),$$

$\xi_{\mathbf{k}} = \omega_{\mathbf{k}}/(\omega_{\mathbf{k}} + \omega_0)$ [7], we obtain

$$H^S = \frac{1}{2}\omega'_0\sigma_z + \sum_{\mathbf{k}} \omega_{\mathbf{k}}a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \sum_{\mathbf{k}} V_{\mathbf{k}}(a_{\mathbf{k}}^\dagger\sigma + \sigma^+a_{\mathbf{k}}), \quad (4)$$

where

$$\omega'_0 = \omega_0 - \frac{d^2}{6\pi^2\epsilon_0 c^3} \int_0^{\omega_c} \frac{2\omega_0\omega_k^3}{(\omega_k + \omega_0)^2} d\omega_k$$

is the transformed transition frequency including the self-energy [7] and $\omega_c = m_e c^2$ is the cutoff frequency with m_e the electron mass, and $V_{\mathbf{k}} = 2\omega_0 g_{\mathbf{k}}/(\omega_0 + \omega_{\mathbf{k}})$ is the effective coupling strength based on the ground state of the whole atom-vacuum system [1,7]. The difference between $V_{\mathbf{k}}$ and $g_{\mathbf{k}}$, $2\omega_0/(\omega_0 + \omega_{\mathbf{k}})$, is resulted from the counterrotating terms. Note that the renormalization for the self-energy is troublesome in the study with the simplified system of two-level atoms because the Thomas-Reiche-Kuhn sum rule is not applicable. The transformed energy shift is still divergent because the self-energy introduced by the term $2\pi\int |\mathbf{P}^\perp|^2 d\tau$ and the direct-scattering terms $(e^2/2mc^2)\mathbf{A}^2$ is not yet subtracted [8,9]. However the renormalized energy shift in the polariz-

ability is negligible and we neglect it in our further analysis.

We now apply a coherent field to the system in order to investigate the polarizability, and a new term $-\mathbf{E}_{cf} \cdot \mathbf{d}$ is introduced into the total Hamiltonian to describe the interaction between the atom-vacuum system and the applied field,

$$H_{tot} = H^S + H_{cf} - \mathbf{E}_{cf} \cdot \mathbf{d} = \frac{1}{2}\omega'_0\sigma_z + \sum_{\mathbf{k}} \omega_{\mathbf{k}}a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \sum_{\mathbf{k}} V_{\mathbf{k}}(a_{\mathbf{k}}^\dagger\sigma + \sigma^+a_{\mathbf{k}}) + \sum_{\mathbf{q}} \omega_{\mathbf{q}}b_{\mathbf{q}}^\dagger b_{\mathbf{q}} + \sum_{\mathbf{q}} i\Omega_{\mathbf{q}}(b_{\mathbf{q}}^\dagger - b_{\mathbf{q}})(\sigma^+ + \sigma), \quad (5)$$

where $\Omega_{\mathbf{q}} = \mathbf{d} \cdot \hat{\mathbf{e}}_{\mathbf{q}}(\omega_{\mathbf{q}}/2\epsilon_0 V)^{1/2}$ is the atom-field coupling strength. $b_{\mathbf{q}}$ and $b_{\mathbf{q}}^\dagger$ are the annihilation and creation operator of the coherent field with $\hat{\mathbf{e}}_{\mathbf{q}}$ the unit polarization vector and $\omega_{\mathbf{q}}$ the frequency. Since the interaction Hamiltonian after unitary transformation has the same form as the one under RWA, we can carry on the calculation following the derivation in [3].

By using the commutation relations $[\sigma, \sigma^+] = -\sigma_z$, $[\sigma, \sigma_z] = 2\sigma$, $[a_{\mathbf{k}}, a_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}$, $[b_{\mathbf{q}}, b_{\mathbf{q}'}^\dagger] = \delta_{\mathbf{q}\mathbf{q}'}$, $[\sigma, a_{\mathbf{k}}] = [\sigma, a_{\mathbf{k}}^\dagger] = 0$, $[\sigma, b_{\mathbf{k}}] = [\sigma, b_{\mathbf{k}}^\dagger] = 0$, and $[a_{\mathbf{k}}, b_{\mathbf{k}}] = [a_{\mathbf{k}}, b_{\mathbf{k}}^\dagger] = 0$, where we have assumed that there is no direct interaction between the vacuum mode and coherent mode, we obtain the Heisenberg equations of motion

$$\dot{\sigma} = \frac{1}{i}[\sigma, H_{tot}] = -i\omega'_0\sigma + i\sum_{\mathbf{k}} V_{\mathbf{k}}\sigma_z a_{\mathbf{k}} - \sum_{\mathbf{q}} \Omega_{\mathbf{q}}\sigma_z(b_{\mathbf{q}}^\dagger - b_{\mathbf{q}}), \quad (6a)$$

$$\dot{a}_{\mathbf{k}} = \frac{1}{i}[a_{\mathbf{k}}, H_{tot}] = -i\omega_{\mathbf{k}}a_{\mathbf{k}} - iV_{\mathbf{k}}\sigma, \quad (6b)$$

$$\dot{b}_{\mathbf{q}} = \frac{1}{i}[b_{\mathbf{q}}, H_{tot}] = -i\omega_{\mathbf{q}}b_{\mathbf{q}} + \Omega_{\mathbf{q}}(\sigma^+ + \sigma). \quad (6c)$$

We first integrate Eqs. (6b) and (6c) and the resulting equations are

$$a_{\mathbf{k}}(t) = a_{\mathbf{k}}(0)e^{-i\omega_{\mathbf{k}}t} - iV_{\mathbf{k}} \int_0^t dt' \sigma(t')e^{i\omega_{\mathbf{k}}(t'-t)}, \quad (7a)$$

$$b_{\mathbf{q}}(t) = b_{\mathbf{q}}(0)e^{-i\omega_{\mathbf{q}}t} + \Omega_{\mathbf{q}} \int_0^t dt' [\sigma^+(t') + \sigma(t')]e^{i\omega_{\mathbf{q}}(t'-t)}. \quad (7b)$$

Next we substitute Eqs. (7a) and (7b) into Eq. (6a). The resulting equation of motion for $\sigma(t)$ is

$$\begin{aligned} \dot{\sigma}(t) = & -i\omega'_0\sigma(t) - i\sum_{\mathbf{k}} V_{\mathbf{k}}\sigma_z(t)a_{\mathbf{k}}(0)e^{-i\omega_{\mathbf{k}}t} \\ & + \sum_{\mathbf{k}} V_{\mathbf{k}}^2 \int_0^t dt' \sigma_z(t')\sigma(t')e^{i\omega_{\mathbf{k}}(t'-t)} + \sum_{\mathbf{q}} \Omega_{\mathbf{q}}\sigma_z(t) \\ & \times [b_{\mathbf{q}}(0)e^{-i\omega_{\mathbf{q}}t} - b_{\mathbf{q}}^\dagger(0)e^{i\omega_{\mathbf{q}}t}] + \sum_{\mathbf{q}} \Omega_{\mathbf{q}}^2 \left\{ \int_0^t dt' \sigma_z(t') \right. \\ & \left. \times [\sigma^+(t') + \sigma(t')]e^{i\omega_{\mathbf{q}}(t'-t)} - \text{H.c.} \right\}. \quad (8) \end{aligned}$$

Suppose the initial field state is a coherent state with wave vector \mathbf{q}_0 and frequency ω , i.e., the atom is polarized in a coherent state $|\phi_f\rangle$ with $b_{\mathbf{q}}|\phi_f\rangle = \alpha\delta_{\mathbf{q}\mathbf{q}_0}|\phi_f\rangle$ and $a_{\mathbf{k}}|\phi_f\rangle = 0$, the expectation value of electric field is

$$\langle E_{cf} \rangle = i(\omega_q/2\varepsilon_0 V)^{1/2}(\alpha e^{-i\omega t} - \alpha^* e^{i\omega t}) = E \cos \omega t, \quad (9)$$

with E the electric amplitude of the coherent field. We can always get the result on the right-hand side of Eq. (9) by adjusting α with a trivial relative phase.

On taking the expectation value on both sides of Eq. (8), we obtain

$$\begin{aligned} \langle \dot{\sigma}(t) \rangle = & -i\omega'_0 \langle \sigma(t) \rangle - iE\bar{d} \langle \sigma_z(t) \rangle \cos \omega t \\ & + \sum_{\mathbf{k}} V_{\mathbf{k}}^2 \int_0^t dt' \langle \sigma_z(t) \sigma(t') \rangle e^{i\omega_{\mathbf{k}}(t'-t)} \\ & + \Omega_{\mathbf{q}_0}^2 \left\{ \int_0^t dt' \langle \sigma_z(t) [\sigma^+(t') + \sigma(t')] \rangle e^{i\omega(t'-t)} - \text{H.c.} \right\}, \end{aligned} \quad (10)$$

where \bar{d} is the averaged dipole moment with $\bar{d}^2 = d^2/3$.

If the atoms and field are weakly coupled, we expect the atom to be mostly in the lower level with $\langle \sigma_z(t) \rangle = -1$ and we make the Markov approximation $\langle \sigma_z(t) \sigma(t') \rangle \cong \langle \sigma_z(t') \sigma(t') \rangle = -\langle \sigma(t') \rangle$. Furthermore we suppose that the atoms have only negligible influence on the field, which can be regarded as undergoing free evolution, i.e., the second term on the right-hand side of Eq. (6c) can be ignored. That is to say, on the right-hand side of Eq. (10), the fourth term can be neglected compared with the second term. In view of these two approximations, Eq. (10) becomes

$$\begin{aligned} \langle \dot{\sigma}(t) \rangle = & -i\omega'_0 \langle \sigma(t) \rangle + iE\bar{d} \cos \omega t \\ & - \sum_{\mathbf{k}} V_{\mathbf{k}}^2 \int_0^t dt' \langle \sigma(t') \rangle e^{i\omega_{\mathbf{k}}(t'-t)}. \end{aligned} \quad (11)$$

The atom can be well regarded as a forced oscillator and we suppose the solution of Eq. (11) has the form

$$\langle \sigma(t) \rangle = p e^{-i\omega t} + q e^{i\omega t}. \quad (12)$$

It follows, on substituting Eq. (12) into Eq. (11), that

$$\begin{aligned} i(\omega'_0 - \omega) p e^{-i\omega t} + i(\omega'_0 + \omega) q e^{i\omega t} = & \frac{iE\bar{d}}{2} (e^{-i\omega t} + e^{i\omega t}) - \sum_{\mathbf{k}} V_{\mathbf{k}}^2 \int_0^t dt' (p e^{i(\omega_{\mathbf{k}} - \omega)(t'-t)} e^{-i\omega t} + q e^{i(\omega_{\mathbf{k}} + \omega)(t'-t)} e^{i\omega t}) \\ = & \frac{iE\bar{d}}{2} (e^{-i\omega t} + e^{i\omega t}) - \frac{d^2}{6\pi\varepsilon_0 c^3} \int_0^\infty d\omega_{\mathbf{k}} \omega_{\mathbf{k}}^3 \frac{4\omega_0^2}{(\omega_{\mathbf{k}} + \omega_0)^2} \left\{ p \left[\delta(\omega_{\mathbf{k}} - \omega) - \frac{i}{\pi(\omega_{\mathbf{k}} - \omega)} \right] e^{-i\omega t} \right. \\ & \left. + q \left[\delta(\omega_{\mathbf{k}} + \omega) + \frac{1}{\pi(\omega_{\mathbf{k}} + \omega)} \right] e^{i\omega t} \right\} \\ = & \frac{iE\bar{d}}{2} (e^{-i\omega t} + e^{i\omega t}) - [\Gamma_-(\omega) - i\Delta_-(\omega)] p e^{-i\omega t} - [\Gamma_+(\omega) - i\Delta_+(\omega)] q e^{i\omega t}, \end{aligned} \quad (13)$$

where

$$\Gamma_{\pm}(\omega) = \frac{d^2 \omega^3}{6\pi\varepsilon_0 c^3} \frac{4\omega_0^2}{(\omega + \omega_0)^2} \theta(\mp \omega), \quad (14a)$$

$$\Delta_{\pm}(\omega) = \frac{d^2}{6\pi^2 \varepsilon_0 c^3} \wp \int_0^\infty \frac{4\omega_0^2}{(\omega_{\mathbf{k}} + \omega_0)^2} \frac{\omega_{\mathbf{k}}^3 d\omega_{\mathbf{k}}}{\omega_{\mathbf{k}} \pm \omega}. \quad (14b)$$

where $\theta(\mp \omega)$ is the step function with $\theta(\omega) = 1$ if $\omega > 0$ and $\theta(\omega) = 0$ otherwise, and \wp denotes the principle value. Comparing two sides of Eq. (13), we obtain

$$p = \frac{E\bar{d}/2}{\omega'_0 - \omega - \Delta_-(\omega) - i\Gamma_-(\omega)}, \quad (15a)$$

$$q = \frac{E\bar{d}/2}{\omega'_0 + \omega - \Delta_+(\omega) - i\Gamma_+(\omega)}. \quad (15b)$$

The polarization is defined via

$$\begin{aligned} P = \bar{d} \langle \sigma_x \rangle = \bar{d} (\langle \sigma \rangle + \langle \sigma^+ \rangle) = 2\bar{d} \text{Re}[(p + q^*) e^{-i\omega t}] \\ \equiv \text{Re}[\alpha(\omega) E e^{-i\omega t}]. \end{aligned} \quad (16)$$

From Eq. (15) and (16) we get the following expression for the polarizability:

$$\begin{aligned} \alpha(\omega) = \frac{d^2}{3} \left[\frac{1}{\omega'_0 - \omega - \Delta_-(\omega) - i\Gamma_-(\omega)} \right. \\ \left. + \frac{1}{\omega'_0 + \omega - \Delta_+(\omega) + i\Gamma_+(\omega)} \right]. \end{aligned} \quad (17)$$

where we have used the dipole average value $\bar{d}^2 = d^2/3$. We note that, in the denominator of the second term on the right-hand side of Eq. (15), $\Gamma_+(\omega) = 0$ for $\omega > 0$, i.e., there are no damping terms in the antiresonant part of the Kramers-Heisenberg formula, which gives the ‘‘zero’’ prescription.

From the Kramers-Heisenberg scattering theory, the differential cross section for a two-level atom is [5]

$$\frac{d\sigma_{sc}}{d\Omega} = \frac{\omega^4 |\hat{\mathbf{e}}_s \cdot \mathbf{d}|^2 |\hat{\mathbf{e}} \cdot \mathbf{d}|^2}{16\pi^2 \varepsilon_0^2 c^4} \left| \frac{1}{\omega_0 - \omega} + \frac{1}{\omega_0 + \omega} \right|^2, \quad (18)$$

where $\hat{\mathbf{e}}$ and $\hat{\mathbf{e}}_s$ are the unit polarization vectors of the incident and scattered light respectively and Ω is the solid angle. The integration over solid angle Ω and summation over the polarizations of the scattered light yield $\sum_{\text{polarizations}} \int |\hat{\mathbf{e}}_s \cdot \mathbf{d}|^2 d\Omega = (8\pi/3)d^2$, and we average over the orientation of the dipole moments $|\hat{\mathbf{e}} \cdot \mathbf{d}|^2 = (1/3)d^2$. Therefore, after integration of Eq. (18), the total cross section is

$$\frac{\omega}{\varepsilon_0 c} \text{Im}[\alpha(\omega)] = \frac{2d^4 \omega_0^2}{9\pi \varepsilon_0^2 c^4} \frac{\omega^4}{(\omega_0^2 - \omega^2)^2 + 16\omega_0^4/(\omega_0 + \omega)^2 (d^2 \omega^3/6\pi \varepsilon_0 c^3)^2}, \quad (21a)$$

and the scattering cross section

$$\begin{aligned} \frac{\omega^4}{2\pi \varepsilon_0^2 c^4} |\alpha(\omega)|^2 &= \frac{2d^4 \omega^4 \omega_0^2}{9\pi \varepsilon_0^2 c^4} \frac{1 + (d^2 \omega^3/6\pi \varepsilon_0 c^3)^2/(\omega_0 + \omega)^2}{(\omega_0^2 - \omega^2)^2 + 16\omega_0^4/(\omega_0 + \omega)^2 (d^2 \omega^3/6\pi \varepsilon_0 c^3)^2} \approx \frac{2d^4 \omega_0^2}{9\pi \varepsilon_0^2 c^4} \frac{\omega^4}{(\omega_0^2 - \omega^2)^2 + 16\omega_0^4/(\omega_0 + \omega)^2 (d^2 \omega^3/6\pi \varepsilon_0 c^3)^2} \\ &= \frac{\omega}{\varepsilon_0 c} \text{Im}[\alpha(\omega)]. \end{aligned} \quad (21b)$$

In the second line in Eq. (21b), we made an approximation under the perturbative condition $(d^2 \omega^3/6\pi \varepsilon_0 c^3)/(\omega_0 + \omega) \ll 1$, and consequently, we have the optical theorem

$$\frac{\omega^4}{2\pi \varepsilon_0^2 c^4} |\alpha(\omega)|^2 = \frac{\omega}{\varepsilon_0 c} \text{Im}[\alpha(\omega)].$$

In the Rayleigh scattering ($\omega \ll \omega_0$), resonance scattering ($\omega = \omega_0$) and Thomson scattering ($\omega \gg \omega_0$) regions, the approximated values of Eq. (21a) are

$$\frac{\omega}{\varepsilon_0 c} \text{Im}[\alpha(\omega)] \approx \begin{cases} \frac{2d^4 \omega^4}{9\pi \varepsilon_0^2 c^4 \omega_0^2}, & \text{for } \omega \ll \omega_0 \\ \frac{2\pi c^2}{\omega^2}, & \text{for } \omega = \omega_0 \\ \frac{2d^4 \omega_0^2}{9\pi \varepsilon_0^2 c^4}, & \text{for } \omega \gg \omega_0. \end{cases} \quad (22)$$

Equation (22) shows that $(\omega/\varepsilon_0 c) \text{Im}[\alpha(\omega)]$ is proportional to ω^4 for the Rayleigh scattering and independent of ω for Thomson scattering, which are the conclusions from Kramers-Heisenberg scattering equation [5]. Comparing Eq. (22) with Eqs. (19) and (20), we see that the optical theorem can be satisfied for both resonant and nonresonant cases. In order to investigate the effect of the counterrotating terms, we compare Eq. (22) with the result under RWA [3,5],

$$\sigma_{sc}^{nr} = \frac{\omega^4 d^4}{18\pi \varepsilon_0^2 c^4} \left| \frac{1}{\omega_0 - \omega} + \frac{1}{\omega_0 + \omega} \right|^2 = \frac{2\omega^4 d^4 \omega_0^2}{9\pi \varepsilon_0^2 c^4 (\omega_0^2 - \omega^2)^2}, \quad (19)$$

for the nonresonant scattering. For resonant scattering ($\omega = \omega_0$), with the introduction of the damping terms in the denominator in Eq. (19), the total cross section is [5]

$$\sigma_{sc}^{re} = \frac{2\pi c^2}{\omega^2}. \quad (20)$$

It is easy to verify that Eq. (17) satisfies the cross relation $\alpha(-\omega) = \alpha^*(\omega)$ and the optical theorem. From Eq. (17), we have the absorption

$$\frac{\omega}{\varepsilon_0 c} \text{Im}[\alpha(\omega)]_{RWA} = \frac{d^4}{18\pi \varepsilon_0^2 c^4} \frac{\omega^4}{(\omega_0 - \omega)^2 + (d^2 \omega^3/6\pi \varepsilon_0 c^3)^2}. \quad (23)$$

Note that Eqs. (21a) and (23) have the same value for resonant scattering, and they are different for the off-resonance scattering. In order to investigate the effect of the counterrotating terms, we rewrite Eq. (21a) as

$$\frac{\omega}{\varepsilon_0 c} \text{Im}[\alpha(\omega)] = \frac{d^4}{18\pi \varepsilon_0^2 c^4} \frac{\omega^4 f(\omega)}{(\omega_0 - \omega)^2 + f^2(\omega) (d^2 \omega^3/6\pi \varepsilon_0 c^3)^2}. \quad (24)$$

Equation (24) differs from Eq. (23) by an overall interference factor $f(\omega)$ (the square of the ratio between $V_{\mathbf{k}}$ and $g_{\mathbf{k}}$) and also a factor $f^2(\omega)$ in the second term of the denominator. It is instructive to write

$$\begin{aligned} f(\omega) &= \left[\frac{2\omega_0}{(\omega_0 + \omega)} \right]^2 = \left(1 + \frac{\omega_0 - \omega}{\omega_0 + \omega} \right)^2 \\ &= (\omega_0 - \omega)^2 \left(\frac{1}{\omega_0 - \omega} + \frac{1}{\omega_0 + \omega} \right)^2. \end{aligned} \quad (25)$$

The overall interference factor clearly shows the interference between rotating terms and counterrotating terms, which tells us that the counterrotating terms have a constructive contribution for $\omega_0 \gg \omega$ because of $(\omega_0 - \omega)/(\omega_0 + \omega) \approx 1 - (2\omega/\omega_0) \approx 1$ (plus negligible second term in the denominator), and a destructive contribution for $\omega_0 \ll \omega$ because of

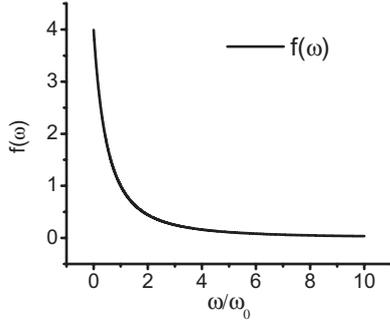


FIG. 1. The ratio of the imaginary part of polarizability without and with RWA. $\text{Im}[\alpha(\omega)]/\text{Im}[\alpha(\omega)]_{RWA} = f(\omega) = 4\omega_0^2/(\omega + \omega_0)^2$.

$(\omega_0 - \omega)/(\omega_0 + \omega) \approx -1 + (2\omega_0/\omega) \approx -1$ (plus negligible second term in the denominator), respectively. At resonance $\omega_0 \approx \omega$, the counterrotating terms have almost no contribution because of $(\omega_0 - \omega)/(\omega_0 + \omega) \approx 0$ and $f(\omega) = 1$.

III. EFFECTS OF THE COUNTERROTATING TERMS

The counterrotating terms of the atom-vacuum Hamiltonian in Eq. (3) brings the correction factor $f(\omega)$ to the decay rate in Eq. (14a) if we neglect the level shift, which gives us hints to take into account the counterrotating terms in a simple way, just by multiplying the damping terms in the result under RWA with $f(\omega)$. For example, the frequency-dependent damping terms, which are obtained by the diagonalization method under RWA in Ref. [5], can be multiplied by $f(\omega)$ to get the same result as Eq. (17).

Before discussing the physical meaning of $f(\omega)$, we show the difference schematically in Fig. 1. The imaginary part of the polarizabilities without and with RWA are different except for the resonance case $\omega = \omega_0$, basically enhanced for low frequency and suppressed for high frequency if we take into account the counterrotating terms.

We now try to interpret the physical meaning of Fig. 1. Equation (12) shows that the atomic dipole is forced to oscillate with the frequency of the applied field ω , rather than its eigenfrequency ω_0 and the atom is therefore dumping energy into the vacuum modes whose frequency is $\omega_k = \omega$, during which the phase evolution due to rotating terms are $\exp[\pm i(\omega_0 - \omega)t]$ while the ones due to counterrotating terms are $\exp[\pm i(\omega_0 + \omega)t]$. For resonance case, the rotating phase evolution is much slower than the counterrotating one, i.e., $|\omega_0 - \omega| \ll \omega_0 + \omega$. In the formulas for transition rates, these factors before time t come into the denominators and because of $1/|\omega_0 - \omega| \gg 1/|\omega_0 + \omega|$, the transition rate is dominated by the rotating-wave terms and so is the imaginary part of polarizability. Therefore, the RWA is legitimate in the resonance case and that is why the polarizability under RWA satisfies the optical theorem in resonance case.

However, for the off-resonance cases, the counterrotating terms are important because the energy cannot be conserved both for the rotating and counterrotating processes. For example, suppose the atom is initially prepared in the ground state, the energy change for the rotating terms, i.e., absorbing a photon and transiting to the excited state, is $\omega_0 - \omega$, while

for the counterrotating terms, emitting a photon and transiting to the excited state, the energy change is $\omega_0 + \omega$; if ω and ω_0 are quite different, we will have $|\omega_0 - \omega| \approx |\omega_0 + \omega|$, consequently, there is no justification for the RWA. We note that these non-energy-conserving transitions can really happen in the presence of the applied field, which acts as the energy source. In the frequency region of the Rayleigh scattering, where $\omega \ll \omega_0$ and $1/(\omega_0 - \omega) \approx 1/(\omega_0 + \omega) \approx 1/\omega_0$, while for the Thomson scattering, where $\omega \gg \omega_0$ and $1/(\omega_0 - \omega) \approx -1/(\omega_0 + \omega) \approx -1/\omega$. The counterrotating terms have nearly the same large contributions as the rotating terms. The two transition amplitudes due to these two processes interfere constructively for the Rayleigh scattering (the same plus sign) but destructively for the Thomson scattering (opposite plus and minus signs). Therefore, after taking into account of the counterrotating terms, we can get a double-transition amplitude for $\omega \ll \omega_0$, four times the transition probability, and also four times of the imaginary part of the polarizability. For $\omega \gg \omega_0$, the transition amplitudes cancel each other, which can be seen in Fig. 1 in the high-frequency region.

IV. FREQUENCY SHIFT

In the denominator of the resonant part of Eq. (15), the shift of the transition frequency is

$$\Delta\omega_0 = \omega'_0 - \omega_0 = -\frac{d^2}{6\pi^2\epsilon_0c^3} \int_0^{\omega_c} \frac{2\omega_0\omega_k^3}{(\omega_k + \omega_0)^2} d\omega_k, \quad (26)$$

which is dependent on the transition frequency but independent of the applied field frequency. The shift of the applied field for the resonant part is

$$\Delta_-(\omega) = \frac{d^2}{6\pi^2\epsilon_0c^3} P \int_0^{\omega_c} \frac{4\omega_0^2}{(\omega_k + \omega_0)^2} \frac{\omega_k^3 d\omega_k}{\omega_k - \omega}, \quad (27)$$

which is dependent on the transition frequency of the atom but also particularly dependent on the field frequency. In the previous studies, the transition shift has been included in the radiative frequency shift, that is to say, both Eqs. (26) and (27) are regarded as the shift of the field. Consequently, the effective radiative frequency shift is

$$\begin{aligned} \delta_-(\omega) &= \Delta_-(\omega) - \Delta\omega_0 \\ &= \frac{d^2}{6\pi^2\epsilon_0c^3} \int_0^{\omega_c} \frac{2\omega_0\omega_k^3}{(\omega_k + \omega_0)^2} \left(1 + \frac{2\omega_0}{\omega_k - \omega}\right) d\omega_k, \end{aligned} \quad (28)$$

which is still different from the results in Refs. [2,3,5], where the counterpart

$$\delta'_-(\omega) = \frac{d^2}{6\pi^2\epsilon_0c^3} \int_0^{\omega_c} \frac{2\omega\omega_k^3}{\omega_k^2 - \omega^2} d\omega_k. \quad (29)$$

We now investigate the effect of this difference on the polarizability in Eq. (17). Because the shift is in the second order of the perturbative coupling strength, which is very small compared with the transition frequency ω_0 , the shift can be neglected for the antiresonant part

$$\frac{1}{\omega'_0 + \omega - \Delta_+(\omega) + i\Gamma_+(\omega)} \approx \frac{1}{\omega_0 + \omega + i\Gamma_+(\omega)}$$

for any positive ω . The resonant part should be treated more carefully, because there is a pole near the real axis. For the off-resonance cases, the detuning between the field frequency and the transition frequency is much larger than the shift, $|\omega - \omega_0| \gg \delta_-(\omega)$, we have

$$\begin{aligned} \frac{1}{\omega'_0 - \omega - \Delta_-(\omega) - i\Gamma_-(\omega)} &\equiv \frac{1}{\omega_0 - \omega - \delta_-(\omega) - i\Gamma_-(\omega)} \\ &= \frac{1}{\omega_0 - \omega - i\Gamma_-(\omega)} \\ &\quad + O\left(\frac{\delta_-(\omega)}{(\omega_0 - \omega)^2}\right). \end{aligned} \quad (30)$$

It does not matter if we replace $\delta_-(\omega)$ with $\delta'_-(\omega)$ in the higher order terms. Therefore, the difference between Eq. (28) and (29) basically has no effect on the polarizability. For the near resonance case, $|\omega - \omega_0| \sim \delta_-(\omega)$, the shift is important. However, under this condition, we have

$$\begin{aligned} \delta_-(\omega) - \delta'_-(\omega) &= \frac{d^2}{6\pi^2\epsilon_0c^3} \int_0^{\omega_c} \frac{2\omega_0\omega_k^3}{(\omega_k + \omega_0)^2} \left(1 + \frac{2\omega_0}{\omega_k - \omega}\right) d\omega_k - \frac{d^2}{6\pi^2\epsilon_0c^3} \int_0^{\omega_c} \frac{2\omega\omega_k^3}{\omega_k^2 - \omega^2} d\omega_k \\ &= \frac{d^2}{6\pi^2\epsilon_0c^3} \int_0^{\omega_c} \frac{2\omega\omega_k^3}{\omega_k^2 - \omega^2} \left[\frac{\omega_0}{\omega} \frac{\omega_k^2 - \omega^2}{(\omega_k + \omega_0)^2} \left(1 + \frac{2\omega_0}{\omega_k - \omega}\right) - 1 \right] d\omega_k \\ &= \frac{d^2}{6\pi^2\epsilon_0c^3} \int_0^{\omega_c} \frac{2\omega\omega_k^3}{\omega_k^2 - \omega^2} \left[\frac{\omega_0(\omega_k + \omega)(\omega_k - \omega + 2\omega_0)}{\omega(\omega_k + \omega_0)^2} - 1 \right] d\omega_k \\ &= \frac{d^2}{6\pi^2\epsilon_0c^3} \int_0^{\omega_c} \frac{2\omega\omega_k^3}{\omega_k^2 - \omega^2} \left\{ \frac{\omega_0 [(\omega_k + \omega_0) + (\omega - \omega_0)] [(\omega_k + \omega_0) - (\omega - \omega_0)]}{\omega(\omega_k + \omega_0)^2} - 1 \right\} d\omega_k \\ &= -\frac{d^2}{6\pi^2\epsilon_0c^3} \int_0^{\omega_c} \frac{2\omega\omega_k^3}{\omega_k^2 - \omega^2} \left[\frac{\omega - \omega_0}{\omega} + \frac{(\omega - \omega_0)^2}{(\omega_k + \omega_0)^2} \right] d\omega_k \sim \frac{d^2}{6\pi^2\epsilon_0c^3} \int_0^{\omega_c} \frac{2\omega\omega_k^3}{\omega_k^2 - \omega^2} \left[\frac{\delta_-(\omega)}{\omega} + \frac{\delta_-^2(\omega)}{\omega_0^2} \right] d\omega_k \\ &= \left[\frac{\delta_-(\omega)}{\omega} + \frac{\delta_-^2(\omega)}{\omega_0^2} \right] \delta'_-(\omega) \approx 0 \end{aligned} \quad (31)$$

from Eq. (28) and (29). Therefore, the polarizability is also the same under perturbative condition in the near resonant case. In conclusion, the difference between Eqs. (28) and (29) is negligible for the polarizability in the full frequency regions.

V. CONCLUDING REMARKS

We calculated the polarizability of a two-level atom using a unitary transformation method. The effects of the counterrotating terms are included into a correction (interference) factor $f(\omega) = 4\omega_0^2/(\omega + \omega_0)^2 = [1 + (\omega_0 - \omega)/(\omega_0 + \omega)]^2$ for the damping rate, and into the shift of the level energy from ω_0 to ω'_0 for the frequency shift of scattered field. In the Rayleigh scattering case $f(\omega)_{\omega \rightarrow 0}$ approaches 4 and in the Thomson scattering case $f(\omega)_{\omega \rightarrow \infty}$ approaches $4\omega_0^2/\omega^2 \rightarrow 0$, which clearly shows the constructive and destructive interference,

respectively, between the resonant (rotating) and antiresonant (counterrotating) terms. Besides, $f(\omega_0) = 1$ ($\omega = \omega_0$) shows the validity of the RWA in the resonant scattering. The obtained result satisfies the cross relation $\alpha(\omega) = \alpha^*(-\omega)$ and the optical theorem. This method greatly simplifies the calculation and has the potential to be extended to multilevel atoms with, for example, a similar unitary transformation plus the density operator methods or the many particle theory [5].

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