Polarizability and the optical theorem for a two-level atom with radiative broadening

Paul R. Berman
Michigan Center for Theoretical Physics, FOCUS Center, and Physics Department, University of Michigan, Ann Arbor, Michigan 48109-1040, USA

Robert W. Boyd
The Institute of Optics, University of Rochester, Rochester, New York 14627, USA

Peter W. Milonni
104 Sierra Vista Dr., Los Alamos, New Mexico 87544, USA
(Received 3 August 2006; published 21 November 2006)

The effect of spontaneous decay on the linear polarizability of an atom is typically included by adding imaginary parts to the frequency denominators that appear in the Kramers-Heisenberg formula. It has been shown for a two-level atom with radiative broadening that these (frequency-dependent) imaginary parts must be included in both the resonant and antiresonant frequency denominators [P. W. Milonni and R. W. Boyd, Phys. Rev. A **69**, 023814 (2004)]; however, the expression obtained by Milonni and Boyd for the polarizability does not satisfy the optical theorem, if contributions from non-rotating-wave terms are included. In this paper, we derive a more accurate expression for the polarizability. The calculations are rather complicated and require that we go beyond the standard Weisskopf-Wigner approximation. We present calculations carried out in both the Heisenberg and Schrödinger pictures, since they offer complementary methods for understanding the dynamics of the Rayleigh scattering associated with the atomic polarizability. Moreover, it is shown that the shifts associated with the excited state are not the Lamb shifts of an isolated atom, but depend on the dynamics of the atom-field interaction. Our results for the polarizability are consistent with those obtained recently by Loudon and Barnett using a Green's-function approach.

DOI: 10.1103/PhysRevA.74.053816 PACS number(s): 42.65.An, 32.10.Dk, 32.70.Jz, 32.80.−t

I. INTRODUCTION

An atom in its ground state has a linear polarizability [1]

$$\alpha(\omega) = \frac{1}{3\hbar}\sum_j |\mathbf{d}_j|^2\left(\frac{1}{\omega_j - \omega} + \frac{1}{\omega_j + \omega}\right) \qquad (1)$$

if it is assumed that the field frequency ω is far removed from any absorption resonance. Here, $\mathbf{d}_j$ and ω_j are the electric dipole matrix element and the angular transition frequency, respectively, connecting the excited state j to the ground state. This (Kramers-Heisenberg) [1] formula ignores the effects of collisions, spontaneous emission, and other line broadening phenomena that can be accounted for by adding imaginary parts to the frequency denominators in Eq. (1), as well as self-energy corrections that appear as additional real terms in both frequency denominators in Eq. (1). Surprisingly enough, however, this familiar procedure is not applied uniformly in the literature. Some authors [2] leave the "antiresonant" denominator $\omega_j+\omega$ unaltered while replacing the "resonant" denominator $\omega_j-\omega$ by $\omega_j-\omega-i\gamma_j$, where γ_j (>0) is the line width of the transition from the ground state to state j. Others advocate a similar change in the antiresonant denominator, but there have been lively debates as to whether the modified denominator should be of the "same-sign" form $\omega_j+\omega-i\gamma_j$ [3] or the "opposite-sign" form $\omega_j+\omega+i\gamma_j$ [4,5]. The causality requirement that $\alpha(\omega)$ should be analytic in the upper half of the complex frequency plane would appear to rule out the same-sign form, and similarly, this form does not satisfy the "crossing relation," $\alpha^*(\omega)=\alpha(-\omega)$, which guarantees that the induced dipole moment is real. However, the situation is more complicated because γ_j is, in general, frequency dependent, as has been emphasized recently in the case of radiative damping, where $\gamma_j \propto \omega^3$ [6]. In that work the radiative damping was found not to affect the antiresonant denominator. Of course, all these correction terms are small if $\gamma_j/|\omega_j \pm \omega| \ll 1$; moreover, in this limit, the underlying atom-field interaction is Rayleigh scattering.

The question of the sign of the damping term in frequency denominators arises also in the case of nonlinear susceptibilities. Long [7], for instance, discusses some historical aspects of this question in the case of Raman scattering and advocates the opposite-sign form.

The fact that antiresonant denominators are at issue in these discussions means, of course, that derivations of the polarizability, including relaxation processes, cannot be based on the usual rotating-wave approximation (RWA). The case of radiative damping is particularly complicated because one must not only go beyond the RWA but must do so within the Weisskopf-Wigner approximation or related approximations that make the calculations tractable. On the other hand, the case of radiative damping provides a simple test of the accuracy of the calculation of $\alpha(\omega)$, namely, that the polarizability satisfies the optical theorem.

Let us briefly recall the form and physical significance of the optical theorem in the context of an atom for which the only mechanism for energy loss in an isotropic environment is radiation. In this case, the optical theorem may be expressed in the form

$$\alpha_I(\omega) = \frac{2\omega^3}{3c^3}|\alpha(\omega)|^2, \tag{2}$$

where $\alpha_I(\omega)$ is the imaginary part of the polarizability. This expression follows from the condition that the rate of change of the atomic (or field) energy is zero. Physically, it simply states that the rate at which the atom's energy increases due to absorption in the presence of a field of frequency ω must exactly balance the rate at which the atom loses energy by radiation, or, equivalently, the rate at which the field energy decreases due to absorption must equal the rate of increase of field energy due to scattering. Thus, the (power) absorption coefficient for the field in a dilute medium of N atoms per unit volume is $a(\omega)=(4\pi\omega/c)N\alpha_I(\omega)$ and, equating this to $N\sigma_R(\omega)$, where

$$\sigma_R(\omega) = \frac{2}{3\pi}\frac{|n-1|^2}{N^2}\left(\frac{\omega}{c}\right)^4 \tag{3}$$

is the cross section for Rayleigh scattering [8] and $n-1 \cong 2\pi N\alpha(\omega)$ is the refractive index, we obtain Eq. (2).

Let us recall also the familiar example of a classical electron oscillator with resonance frequency ω_0. Including the radiation reaction force $2e^2\dddot{x}/3c^3$ in the equation for the electron displacement x in the case of a driving field of amplitude E_0 and frequency ω, we have

$$m\ddot{x} + \omega_0^2 x - \frac{2e^2}{3mc^3}\dddot{x} = \frac{e}{m}E_0 \cos \omega t, \tag{4}$$

and, therefore,

$$\alpha(\omega) = \frac{e^2/m}{\omega_0^2 - \omega^2 - 2ie^2\omega^3/3mc^3}, \tag{5}$$

and the optical theorem (2) is satisfied. Since a two-level atom (TLA) that remains with high probability in its ground state can for many purposes be approximated by such an oscillator in which e^2/m is replaced by $e^2 f/m$, where f is the oscillator strength of the transition, it might be expected that the optical theorem for a TLA follows trivially from (5). We show that things are not so simple.

In light of the recent controversy concerning the same-sign and opposite-sign forms, the polarizability for a two-level atom was recently revisited by Milonni and Boyd [9], hereafter referred to as MB, for the case of radiative damping. It was found that

$$\alpha(\omega) = \frac{d^2/\hbar}{\omega_0 - \omega - \delta(\omega) - i\gamma(\omega)} + \frac{d^2/\hbar}{\omega_0 + \omega + \delta(\omega) + i\gamma(\omega)}, \tag{6}$$

where d is the TLA transition dipole moment, which may be taken to be real by an appropriate choice of the phases of the lower- and upper-state wave functions, ω_0 is the TLA transition frequency, and

$$\delta(\omega) = \Delta_-(\omega) - \Delta_+(\omega), \tag{7}$$

$$\Delta_\pm(\omega) = \frac{2d^2}{3\pi\hbar c^3}\mathrm{P}\int_0^\infty \frac{d\Omega\Omega^3}{\Omega \pm \omega}, \tag{8}$$

$$\gamma(\omega) = \Gamma_-(\omega) + \Gamma_+(\omega), \tag{9}$$

P denotes principal part and

$$\Gamma_\pm(\omega) = \frac{2d^2}{3\hbar c^3}\int_0^\infty d\Omega\Omega^3\delta(\Omega \pm \omega) \tag{10}$$

such that

$$\Gamma_+(\omega) = 0; \quad \Gamma_-(\omega) = 2d^2\omega^3/3\hbar c^3$$

for $\omega > 0$. These results were obtained using the electric dipole form of the atom interaction with the quantized field. The divergent radiative level shifts $\Delta_\pm(\omega)$ must obviously be renormalized, but this was not of direct concern in MB [9] nor is it of concern here; the (mass) renormalization process in any event cannot be correctly carried out in the two-level approximation, since the dipole sum rule does not hold in the restricted Hilbert space of the TLA. We can write

$$\delta(\omega) = -[\Delta E_2(\omega) - \Delta E_1(\omega)]/\hbar, \tag{11}$$

where $\Delta E_2(\omega) = -\hbar\Delta_-(\omega)$ and $\Delta E_1(\omega) = -\hbar\Delta_+(\omega)$ are the level shifts of the upper ($|2\rangle$) and lower ($|1\rangle$) states, respectively, of the TLA. $\Delta E_2(\omega_0)$ and $\Delta E_1(\omega_0)$, when modified to account for the fact that we have used the electric dipole rather than minimal coupling form of the atom-field interaction, are the nonrelativistic, unrenormalized "Lamb shifts" of the TLA in the absence of an applied field [10]. Note that both the transition width and shift in (6) are evaluated at the applied field frequency rather than the atomic transition frequency.

The purpose of MB [9] was to address the question of the damping term in the antiresonant denominator in the specific case of radiative broadening. The main conclusions to be drawn from the expression (6) are that (i) the radiative damping rate appears in both the resonant and the antiresonant denominators, and (ii) this damping rate is consistent with the opposite-sign convention, albeit the rate is, in fact, frequency dependent. The expression (6) can be rewritten, assuming $\gamma^2(\omega) \ll |\omega_0^2 - \omega^2|$ and ignoring for the moment the frequency shift $\delta(\omega)$, as [11]

$$\alpha(\omega) = \frac{2\omega_0 d^2/\hbar}{\omega_0^2 - \omega^2 - 2i\omega\gamma(\omega)} = \frac{2\omega_0 d^2/\hbar}{\omega_0^2 - \omega^2 - 2id^2\omega^4/3\hbar c^3}. \tag{12}$$

The factor ω^4 in the damping term in the denominator contrasts with the ω^3 in the classical expression (5). It follows easily from Eq. (2) that a consequence of this difference is that (12) does *not* satisfy the optical theorem.

The major goal of this paper is to obtain a better approximation than (6) for the TLA polarizability in the case of radiative broadening. The results we obtain satisfy the optical theorem within the order of validity of the approximation. Although the TLA polarizability of interest here can approximate that of a real atom in the case of a single ground-to-excited-state transition with large oscillator strength, our primary interest here is in the calculation itself, which, as noted earlier, requires one to go beyond a Weisskopf-Wigner-type approximation, which in its standard form involves the rotating-wave approximation as well as the

Markov approximation for which the line width and shift are frequency dependent. To gain additional insight into the dynamics responsible for the polarizability, we present calculations using both the Heisenberg and Schrödinger pictures. This is a rare case in quantum optics in which a Schrödinger-picture calculation is no more difficult than a Heisenberg-picture one [13]; moreover, the Schrödinger-picture calculation is "exact" within certain limits, whereas the Heisenberg-picture approach is perturbative. It must be mentioned that a different approach based on time-dependent Green's functions has been taken by Loudon and Barnett [12], who first obtained corrections to (6) that allow the optical theorem to be satisfied. Following the calculations of the following three sections, we compare the assumptions and approximations made in these different approaches.

An important feature of the calculation is that it allows one to investigate the origin of the level shifts that enter. The Lamb shifts of the levels of an isolated atom can be calculated in an unambiguous way; however, the level shifts that appear in our calculation of the polarizability cannot be interpreted solely in terms of these Lamb shifts, since the shifts associated with the excited state that we find depend on the dynamics of the atom-field interaction and are not the Lamb shifts of an isolated atom [14]. This point is discussed in more detail at the end of Sec. IV.

II. HEISENBERG PICTURE I

We use essentially the same Hamiltonian and notation as MB [9] for the interaction of a two-level atom with the field in the electric dipole approximation, except that we treat the applied field classically from the outset. For an applied field $E_0 \cos \omega t$ polarized along the x direction, the Hamiltonian is then

$$H = \tfrac{1}{2}\hbar\,\omega_0\sigma_z + \hbar\,\omega_k a_k^\dagger a_k - dE_0 \cos \omega t(\sigma + \sigma^\dagger) - i\hbar\,C_k(\sigma a_k + \sigma^\dagger a_k - a_k^\dagger \sigma - a_k^\dagger \sigma^\dagger). \qquad (13)$$

a_k and $a_k^\dagger$ are as usual the free-field photon annihilation and creation operators for mode k, where k for brevity denotes the wave vector $\mathbf{k}$ and the polarization index λ $(=1,2)$ of a free-space, plane-wave field mode, and σ, $\sigma^\dagger$, and σ_z are, respectively, the TLA lowering, raising, and population difference operators. We use a summation convention in which repeated field indices are to be summed over on the right-hand side of an equation unless they appear explicitly on the left-hand side. The coupling constant $C_k \equiv (\mathbf{d}\cdot\mathbf{e}_{\mathbf{k}\lambda}) \times (2\pi\omega_k/\hbar V)^{1/2}$, where $\mathbf{e}_{\mathbf{k}\lambda}$ is the polarization unit vector for the mode $(\mathbf{k},\lambda)$ and V is the quantization volume. Without loss of generality for our purposes, we take C_k to be real and positive. As in MB [9], we write the Heisenberg equation of motion for $\sigma(t)$ and take expectation values over an initial state in which the initial TLA state is the lower state and the initial field state is that in which all modes are unoccupied except for that corresponding to the external, classically described field

$$\langle\dot{\sigma}(t)\rangle = -i\omega_0\langle\sigma(t)\rangle - i\frac{d}{\hbar}E_0 \cos \omega t\langle\sigma_z(t)\rangle + C_k^2\int_0^t dt'[\langle\sigma_z(t)\sigma(t')\rangle + \langle\sigma_z(t)\sigma^\dagger(t')\rangle]e^{i\omega_k(t'-t)} - C_k^2\int_0^t dt'[\langle\sigma^\dagger(t')\sigma_z(t)\rangle + \langle\sigma(t')\sigma_z(t)\rangle]e^{-i\omega_k(t'-t)}. \qquad (14)$$

Since the TLA is assumed to be initially in the lower state, we approximate $\langle\sigma_z(t)\rangle$ in the second term on the right-hand side of (14) by -1; this is the familiar approximation in which the atom responds to the field as a classical Lorentz oscillator but with the factor e^2/m replaced by e^2f/m, where $f = 2m\omega_0 d^2/e^2\hbar$ is the oscillator strength. This approximation assumes that the applied field frequency ω is far enough removed from the absorption resonance that the atom remains with high probability in the lower state. This approximation is made in MB [9], together with the "Markovian approximation" in the form

$$\langle\sigma_z(t)\sigma(t')\rangle \cong \langle\sigma_z(t')\sigma(t')\rangle = -\langle\sigma(t')\rangle \qquad (15)$$

and likewise for the remaining three terms in the integrals on the right-hand side of (14). These approximations lead straightforwardly to the polarizability (6).

Here, we improve on the Markovian approximation (15) by using in (14) the formal solution of the Heisenberg equation of motion for $\sigma_z(t)$ that follows from the Hamiltonian (13):

$$\sigma_z(t) = \sigma_z(t') + 2i\frac{dE_0}{\hbar}\cos \omega t\int_{t'}^t dt''[\sigma(t'') - \sigma^\dagger(t'')] + 2C_k\int_{t'}^t dt''\{[\sigma(t'') - \sigma^\dagger(t'')]a_k(t'') - a_k^\dagger(t'')[\sigma(t'') - \sigma^\dagger(t'')]\}. \qquad (16)$$

Consider the first term in the first integral on the right-hand side of (14) when (16) and the equal-time operator identity $\sigma_z(t')\sigma(t') = -\sigma(t')$ are used. We ignore terms of third- and higher-order in the atom field coupling C_k, which amounts to dropping the third term in (16) when (16) is used in (14). In this approximation, which we discuss in more detail later,

$$C_k^2\int_0^t dt'\langle\sigma_z(t)\sigma(t')\rangle e^{i\omega_k(t'-t)} \cong -C_k^2\int_0^t dt'\langle\sigma(t')\rangle e^{i\omega_k(t'-t)} - \frac{id}{\hbar}E_0C_k^2\int_0^t dt' e^{i\omega_k(t'-t)}\int_{t'}^t dt''[e^{-i\omega t''} + e^{i\omega t''}] \times[\langle\sigma(t'')\sigma(t')\rangle - \langle\sigma^\dagger(t'')\sigma(t')\rangle]. \qquad (17)$$

To remain to second order in the atom-field coupling, we make the replacements

$$\langle \sigma(t'')\sigma(t')\rangle \to \langle \sigma(0)\sigma(0)\rangle e^{-i\omega_0(t''+t')} = 0,$$

$$\langle \sigma^\dagger(t'')\sigma(t')\rangle \to \langle \sigma^\dagger(0)\sigma(0)\rangle e^{i\omega_0(t''-t')} = 0. \tag{18}$$

The first expression is identically zero, whereas the second is zero under the assumption that the initial TLA state is the lower state $|1\rangle$ ($\sigma(0)|1\rangle=0$). Thus,

$$C_k^2\int_0^t dt'\langle \sigma_z(t)\sigma(t')\rangle e^{i\omega_k(t'-t)} \cong -C_k^2\int_0^t dt'\langle \sigma(t')\rangle e^{i\omega_k(t'-t)}, \tag{19}$$

which is equivalent to the approximation (15) made in MB [9] for the first term in the first integral on the right-hand side of (14).

Consider next the second term in the first integral on the right-hand side of (14) when (16) and the identity $\sigma_z(t')\sigma^\dagger(t')=\sigma^\dagger(t')$ are used. Following the same approximation leading to (17), we make the replacement

$$\begin{aligned}&C_k^2\int_0^t dt'\langle \sigma_z(t)\sigma^\dagger(t')\rangle e^{i\omega_k(t'-t)}\\ &\to \sum_k C_k^2\int_0^t dt'\langle \sigma^\dagger(t')\rangle e^{i\omega_k(t'-t)}\\ &- \frac{id}{\hbar}E_0C_k^2\int_0^t dt' e^{i\omega_k(t'-t)}\int_{t'}^t dt''[e^{-i\omega t''}+e^{i\omega t''}]\\ &\times\langle[\sigma(t'')\sigma^\dagger(t')-\sigma^\dagger(t'')\sigma^\dagger(t')]\rangle.\end{aligned} \tag{20}$$

As in (18), we take

$$\langle \sigma(t'')\sigma^\dagger(t')\rangle \cong \langle \sigma(0)\sigma^\dagger(0)\rangle e^{-i\omega_0(t''-t')} = e^{-i\omega_0(t''-t')},$$

$$\langle \sigma^\dagger(t'')\sigma^\dagger(t')\rangle \cong \langle \sigma^\dagger(0)\sigma^\dagger(0)\rangle e^{i\omega_0(t''+t')} = 0 \tag{21}$$

for the initial TLA state $|1\rangle$. Then,

$$\begin{aligned}&C_k^2\int_0^t dt'\langle \sigma_z(t)\sigma^\dagger(t')\rangle e^{i\omega_k(t'-t)}\\ &\cong C_k^2\int_0^t dt'\langle \sigma^\dagger(t')\rangle e^{i\omega_k(t'-t)} - \frac{id}{\hbar}E_0C_k^2\int_0^t dt' e^{i\omega_k(t'-t)}\int_{t'}^t dt''\\ &\times[e^{-i\omega t''}+e^{i\omega t''}]e^{-i\omega_0(t''-t')}\\ &= C_k^2\int_0^t dt'\langle \sigma^\dagger(t')\rangle e^{i\omega_k(t'-t)} + \frac{d}{\hbar}E_0\\ &\times\frac{1}{\omega+\omega_0}e^{-i\omega t}C_k^2\int_0^t dt'[e^{i(\omega_k+\omega_0)(t'-t)}-e^{i(\omega_k-\omega)(t'-t)}]\\ &-\frac{d}{\hbar}E_0\frac{1}{\omega-\omega_0}e^{i\omega t}C_k^2\int_0^t dt'[e^{i(\omega_k+\omega_0)(t'-t)}-e^{i(\omega_k+\omega)(t'-t)}].\end{aligned} \tag{22}$$

For the times $t \gg 1/\omega$ of interest, we use

$$C_k^2\int_0^t dt' e^{i(\omega_k\pm\omega)(t'-t)} = \Gamma_\pm(\omega) - i\Delta_\pm(\omega). \tag{23}$$

(We depart here from the notation in MB [9] by using $\Gamma_\pm(\omega)$ instead of $\gamma_\pm(\omega)$ and using $\gamma(\omega)$ to denote $\Gamma_+(\omega)+\Gamma_-(\omega)$ [Eq. (9)]. Then, (22) becomes

$$\begin{aligned}&C_k^2\int_0^t dt'\langle \sigma_z(t)\sigma^\dagger(t')\rangle e^{i\omega_k(t'-t)}\\ &\cong C_k^2\int_0^t dt'\langle \sigma^\dagger(t')\rangle e^{i\omega_k(t'-t)} + \frac{dE_0}{\hbar}\frac{e^{-i\omega t}}{\omega+\omega_0}\\ &\times[\Gamma_+(\omega_0)-\Gamma_-(\omega)-i\Delta_+(\omega_0)+i\Delta_-(\omega)]\\ &-\frac{dE_0}{\hbar}\frac{e^{i\omega t}}{\omega-\omega_0}[\Gamma_+(\omega_0)-\Gamma_+(\omega)-i\Delta_+(\omega_0)+i\Delta_+(\omega)].\end{aligned} \tag{24}$$

We proceed in the same way to evaluate, approximately, the second integral on the right-hand side of (14). Collecting all the terms, we obtain the following approximate equation for $\langle \sigma(t)\rangle$:

$$\begin{aligned}\langle \dot\sigma(t)\rangle &= -i\omega_0\langle \sigma(t)\rangle + \frac{id}{2\hbar}E_0[e^{-i\omega t}+e^{i\omega t}]\\ &+ C_k^2\int_0^t dt'[-\langle \sigma(t')\rangle + \langle \sigma^\dagger(t')\rangle]e^{i\omega_k(t'-t)}\\ &- C_k^2\int_0^t dt'[-\langle \sigma^\dagger(t')\rangle + \langle \sigma(t')\rangle]e^{-i\omega_k(t'-t)}\\ &+ \frac{d}{\hbar}E_0e^{-i\omega t}\left[\frac{-\Gamma_-(\omega)+i\Delta_-(\omega)-i\Delta_+(\omega_0)}{\omega+\omega_0}\right.\\ &\left.+\frac{-\Gamma_+(\omega)+i\Delta_+(\omega_0)-i\Delta_+(\omega)}{\omega-\omega_0}\right]\\ &- \frac{d}{\hbar}E_0e^{i\omega t}\left[\frac{-\Gamma_-(\omega)+i\Delta_+(\omega_0)-i\Delta_-(\omega)}{\omega+\omega_0}\right.\\ &\left.+\frac{-\Gamma_+(\omega)+i\Delta_+(\omega)-i\Delta_+(\omega_0)}{\omega-\omega_0}\right],\end{aligned} \tag{25}$$

where we have used the fact that $\Gamma_+(\omega_0)=0$.

The solution of (25) has the form $\langle \sigma(t)\rangle = se^{-i\omega t}+re^{i\omega t}$, and the induced dipole moment $p=d\langle \sigma+\sigma^\dagger\rangle = 2d\,\mathrm{Re}[(s+r^*)e^{-i\omega t}] \equiv \mathrm{Re}[\alpha(\omega)E_0e^{-i\omega t}]$. Solving (25) for s and r, we obtain

$$\begin{aligned}\alpha(\omega) &\cong \frac{2d^2\omega_0}{\hbar}\frac{1}{\omega_0^2-\omega^2-2i\omega[\gamma(\omega)-i\delta(\omega)]}\\ &\times\left[1+2\frac{\Delta_-(\omega)+i\Gamma_-(\omega)-\Delta_+(\omega_0)}{\omega+\omega_0}\right.\\ &\left.+2\frac{\Delta_+(\omega_0)-\Delta_+(\omega)+i\Gamma_+(\omega)}{\omega-\omega_0}\right]\end{aligned} \tag{26}$$

for the TLA polarizability. This expression satisfies the crossing relation $\alpha^*(-\omega)=\alpha(\omega)$. If we retain only terms up to fourth order in d (recall that all the widths and shifts are of order d^2), consistent with the approximations used in our derivation, then the optical theorem (2) is also satisfied to this order. We defer further discussion of this result to Sec. V.

In this section, we have shown how to generalize the results of MB [9] to obtain an expression for the polarizability that is consistent with the optical theorem; however, it is difficult to rigorously justify the factorization approximations that were used in arriving at this result. We now give alternative derivations based a Schrödinger-picture approach and a Heisenberg picture, in which the approximations introduced are more transparent. Moreover, these alternative derivations provide additional insight into the underlying physical processes contributing to the polarizability.

III. SCHRÖDINGER PICTURE

In many cases, it is easier, computationally, to evaluate expectation values of atomic Heisenberg operators, such as the polarizability, using a Heisenberg- rather than Schrödinger-picture approach. In this case, however, since we are working to first order in the external field amplitude, the Schrödinger approach is no more difficult than the Heisenberg approach. Moreover, it is a simple matter to keep track of the amplitudes that contribute and no factorization approximations are needed. Within certain approximations to be specified below, this approach is exact.

The state vector for the system can be written as

$$|\psi\rangle = b_{1,0}|1,0\rangle + b_{2,0}|2,0\rangle + b_{1,k}|1,k\rangle + b_{2,k}|2,k\rangle + b_{1,kk'}|1,kk'\rangle + b_{2,kk'}|2,kk'\rangle + \cdots . \tag{27}$$

The field states, which are specified by the second label in each ket, refer to vacuum field modes; as before, the externally applied field is treated classically. Thus, $|1,0\rangle$ and $|2,0\rangle$ are, respectively, the states in which the TLA is in the lower state and the upper state and the field state is the vacuum. $|1,k\rangle$ and $|2,k\rangle$ are states in which the TLA is in the lower state and the upper state, respectively, and there is a single photon in the field mode denoted by k, as in the preceding section. Similarly, $|1,kk'\rangle$ and $|2,kk'\rangle$ are states in which the TLA is in the lower state and the upper state, respectively, and there is a photon in mode k and a photon in mode k'.

The expectation value of the electric dipole moment is

$$p = p_+e^{-i\omega t} + p_-e^{i\omega t} = d(\rho_{21} + \rho_{12}), \tag{28}$$

where $\rho_{21}=\rho_{12}^*$ is an off-diagonal density matrix element. Writing

$$\rho_{21} = \rho_{21}^+e^{-i\omega t} + \rho_{21}^-e^{i\omega t}, \tag{29}$$

we can express the complex polarizability as

$$\alpha = \frac{p_+}{E_+} = \frac{d^2\beta}{\hbar}, \tag{30}$$

where

$$\beta = \frac{\lim_{\chi\to 0}[\rho_{21}^+ + (\rho_{21}^-)^*]}{\chi} \tag{31}$$

and the Rabi frequency is defined by $\chi = dE_0/2\hbar$.

The total (cycle-averaged) power radiated by the TLA is

$$P = \frac{\omega^4}{3c^3}|\alpha|^2E_0^2 = \frac{4d^2\omega^4\chi^2|\beta|^2}{3c^3}, \tag{32}$$

and the rate at which energy is lost from the external field is calculated straightforwardly to be

$$P = 2\hbar\omega\chi^2\,\mathrm{Im}[\beta]. \tag{33}$$

The optical theorem is satisfied if these two expressions are equal

$$\mathrm{Im}[\beta] = \frac{2\omega^3d^2}{3\hbar c^3}|\beta|^2 = \Gamma_-(\omega)|\beta|^2. \tag{34}$$

In order to obtain β, one is faced with the task of calculating

$$\rho_{21} = \rho_{2,0;1,0} + \rho_{2,k;1,k} + \rho_{2,kk';1,kk'} + \cdots \tag{35}$$

and extracting ρ_{21}^+ and ρ_{21}^- from this quantity. To first order in χ, it turns out that terms beyond the first two make contributions to ρ_{21}^+ and ρ_{21}^- of order $C_k^2/(\omega_0+\omega_k)^2$, that is, of order of the TLA "Lamb shift" divided by ω_0. Such terms, which also determine the (non-RWA) excited-state population in the absence of any applied fields, are systematically neglected in this work, as are terms of order $C_k^2/(\omega_0+\omega_k)(\omega+\omega_k)$:

$$\frac{C_k^2}{(\omega_0+\omega_k)^2} \ll 1, \tag{36a}$$

$$\frac{C_k^2}{(\omega_0+\omega_k)(\omega+\omega_k)} \ll 1. \tag{36b}$$

As a consequence, the density matrix element (35) can be approximated for our purposes as

$$\rho_{21} \cong \rho_{2,0;1,0} + \rho_{2,k;1,k}. \tag{37}$$

Instead of directly calculating density matrix elements, we calculate probability *amplitudes* and form density matrix elements from these amplitudes. This simplifies the calculation considerably and enables us to obtain an expression for $\beta(\omega)$ that is essentially *exact* to all orders in the vacuum coupling strength. An amplitude approach can be used in this problem since the vacuum field states responsible for relaxation are included explicitly in the state amplitudes. Such a procedure is *practical* in our case owing to the fact that the calculation is perturbative in the applied field—for a strong external field (Rabi frequency greater than decay rates), the number of terms that enter would render the amplitude approach virtually useless.

Owing to the definition (31), we need only calculate ρ_{21} to first order in χ. The equations for the probability amplitudes are easily obtained from the Hamiltonian (13) and the time-dependent Schrödinger equation [15]:

$$\dot{b}_{1,0} = i\chi(e^{i\omega t} + e^{-i\omega t})b_{2,0} - C_kb_{2,k}, \tag{38a}$$

$$\dot{b}_{2,0} = i\chi(e^{i\omega t} + e^{-i\omega t})b_{1,0} - i\omega_0b_{2,0} - C_kb_{1,k}, \tag{38b}$$

$$\dot{b}_{1,k} = C_k b_{2,0} - C_{k'} b_{2,kk'} - i\omega_k b_{1,k} + i\chi(e^{i\omega t} + e^{-i\omega t}) b_{2,k}, \tag{38c}$$

$$\dot{b}_{2,k} = i\chi(e^{i\omega t} + e^{-i\omega t}) b_{1,k} - i(\omega_0 + \omega_k) b_{2,k} + C_k b_{1,0}, \tag{38d}$$

$$\dot{b}_{2,kk'} = -i(\omega_0 + \omega_k + \omega_{k'}) b_{2,kk'} + C_{k'} b_{1,k} + C_k b_{1,k'}. \tag{38e}$$

We neglect ground-state amplitudes involving two or more free-field photons and excited-state amplitudes involving three or more free-field photons, since they lead to corrections of order (36); the probability amplitudes we retain are the only ones that contribute to ρ_{21} to first order in the external field. These probability amplitudes satisfy Eqs. (38), which are to be solved to first order in χ and to *all* orders in the vacuum coupling C_k.

We are interested in steady-state solutions for the probability amplitudes. From the structure of Eqs. (38), we deduce that $b_{2,0}$, $b_{1,k}$, and $b_{2,kk'}$ oscillate as $e^{\pm i\omega t}$ and depend linearly on χ, while $b_{1,0}$ and $b_{2,k}$ have no linear dependence on χ. To first order in χ, then, the solution of (38d) is

$$b_{2,k} \cong \frac{-iC_k b_{1,0}}{\omega_0 + \omega_k}, \tag{39}$$

and, substituting this back into (38a) (and neglecting the lead term, which is of order χ^2), we obtain

$$\dot{b}_{1,0} \cong i\Delta_+(\omega_0) b_{1,0}. \tag{40}$$

In obtaining a solution to Eqs. (38) and (40) to first order in χ, we find that $b_{2,0}$, $b_{1,k}$, and $b_{2,kk'}$ oscillate as $e^{\pm i\omega t + i\Delta_+(\omega_0)t}$ while $b_{1,0}$ and $b_{2,k}$ oscillate as $e^{i\Delta_+(\omega_0)t}$. Thus, we assume a trial solution of the form

$$b_{1,0} = e^{i\Delta_+(\omega_0)t} \tag{41a}$$

$$b_{2,0} = (b_{2,0}^{+} e^{-i\omega t} + b_{2,0}^{-} e^{i\omega t}) e^{i\Delta_+(\omega_0)t} \tag{41b}$$

$$b_{1,k} = (b_{1,k}^{+} e^{-i\omega t} + b_{1,k}^{-} e^{i\omega t}) e^{i\Delta_+(\omega_0)t} \tag{41c}$$

$$b_{2,k} = \tilde{b}_{2,k} e^{i\Delta_+(\omega_0)t} = \frac{-iC_k}{[\omega_0 + \omega_k + \Delta_+(\omega_0)]} e^{i\Delta_+(\omega_0)t} \tag{41d}$$

$$b_{2,kk'} = (b_{2,kk'}^{+} e^{-i\omega t} + b_{2,kk'}^{-} e^{i\omega t}) e^{i\Delta_+(\omega_0)t}. \tag{41e}$$

Note that $|b_{1,0}| = 1$ and $|b_{2,k}| = C_k / [\omega_0 + \omega_k + \Delta_+(\omega_0)]$ in the approximations implicit in (41); the fact that $|b_{2,k}| \neq 0$ results from fluctuations of the vacuum field that lead to a small steady-state value for this amplitude (but not to significant population or to measurable radiation in the absence of any applied field).

When we substitute our trial solution into Eqs. (38), and keep terms only of first order in χ, we obtain the steady-state equations

$$0 = i\chi - i[\Delta + \Delta_+(\omega_0)] b_{2,0}^{+} - C_k b_{1,k}^{+} \tag{42a}$$

$$0 = i\chi - i[\Delta^{(+)} + \Delta_+(\omega_0)] b_{2,0}^{-} - C_k b_{1,k}^{-} \tag{42b}$$

$$0 = C_k b_{2,0}^{+} - C_{k'} b_{2,kk'}^{+} - i[\omega_k - \omega + \Delta_+(\omega_0) - i\epsilon] b_{1,k}^{+} + i\chi \tilde{b}_{2,k} \text{ (no sum on } k) \tag{42c}$$

$$0 = C_k b_{2,0}^{-} - C_{k'} b_{2,kk'}^{-} - i[\omega_k + \omega + \Delta_+(\omega_0)] b_{1,k}^{-} + i\chi \tilde{b}_{2,k} \text{ (no sum on } k) \tag{42d}$$

$$0 = -i[\omega_0 - \omega + \omega_k + \omega_{k'} + \Delta_+(\omega_0)] b_{2,kk'}^{+} + C_{k'} b_{1,k}^{+} + C_k b_{1,k'}^{+} \text{ (no sum)} \tag{42e}$$

$$0 = -i[\omega_0 + \omega + \omega_k + \omega_{k'} + \Delta_+(\omega_0)] b_{2,kk'}^{-} + C_{k'} b_{1,k}^{-} + C_k b_{1,k'}^{-} \text{ (no sum)}, \tag{42f}$$

where

$$\Delta = \omega_0 - \omega; \quad \Delta^{(+)} = \omega_0 + \omega, \tag{43}$$

$$\tilde{b}_{2,k} \approx \frac{-iC_k}{\omega_0 + \omega_k}. \tag{44}$$

The frequency ω_k has been replaced by $\omega_k - i\epsilon$ in Eq. (42c), where ϵ is a positive quantity that tends toward zero, to ensure that a steady-state solution for $b_{1,k}^{+}$ exists. In terms of these parameters, it follows from Eqs. (29), (31), (37), and (41), and the fact that $\rho_{a,a'} = b_\alpha b_{\alpha'}^{*}$, that

$$\beta = \frac{b_{2,0}^{+} + (b_{2,0}^{-})^{*} + \tilde{b}_{2,k}(b_{1,k}^{-})^{*} + \tilde{b}_{2,k}^{*} b_{1,k}^{+}}{\chi}. \tag{45}$$

Equations (42) break up into two uncoupled sets of equations, one for $b_{2,0}^{+}$, $b_{1,k}^{+}$, $\tilde{b}_{2,k}$, $b_{2,kk'}^{+}$ and the other for $b_{2,0}^{-}$, $b_{1,k}^{-}$, $\tilde{b}_{2,k}$, $b_{2,kk'}^{-}$. However, it is sufficient to solve the first set only since Eqs. (42) imply that

$$b_{a,a'}^{-}(\omega) = b_{a,a'}^{+}(-\omega)$$

$$\tilde{b}_{2,k}(\omega) = \tilde{b}_{2,k}(-\omega) \tag{46}$$

for any $\{a, a'\}$. Therefore,

$$\beta(\omega) = \frac{b_{2,0}^{+}(\omega) + [b_{2,0}^{-}(\omega)]^{*} + \tilde{b}_{2,k}(\omega)[b_{1,k}^{-}(\omega)]^{*} + \tilde{b}_{2,k}^{*}(\omega)b_{1,k}^{+}(\omega)}{\chi}$$
$$= \frac{b_{2,0}^{-}(-\omega) + [b_{2,0}^{+}(-\omega)]^{*} + \tilde{b}_{2,k}(-\omega)[b_{1,k}^{+}(-\omega)]^{*} + \tilde{b}_{2,k}^{*}(-\omega)b_{1,k}^{-}(-\omega)}{\chi} = \beta^{*}(-\omega),$$

i.e., the crossing relation is satisfied.

We now solve Eqs. (42) by eliminating $b_{2,0}^{+}$, $\tilde{b}_{2,k}$, and $b_{2,kk'}^{+}$ to obtain an (integral) equation for $b_{1,k}^{+}$ that is solved exactly using a Born series. We already have $\tilde{b}_{2,k}$ from (44), and we can use (42e) to eliminate $b_{2,kk'}^{+}$

$$b_{2,kk'}^{+} = \frac{C_{k'}b_{1,k}^{+} + C_k b_{1,k'}^{+}}{\omega_0 - \omega + \omega_k + \omega_{k'}}. \tag{47}$$

[In writing (44) and (47), and in what follows, we neglect $\Delta_{+}(\omega_0)$ in all frequency denominators *except* those involving Δ and $\omega_k-\omega$, since it represents a correction of order (36).] Substituting this into (42c), and using (44), we obtain

$$\begin{aligned} 0 = C_k b_{2,0}^{+} &+ i\frac{C_{k'}^2 b_{1,k}^{+} + C_{k'}C_k b_{1,k'}^{+}}{\omega_0 - \omega + \omega_k + \omega_{k'}} \\ &- i[\omega_k - \omega + \Delta_{+}(\omega_0) - i\epsilon]b_{1,k}^{+} \\ &+ \chi\frac{C_k}{\omega_0 + \omega_k} \text{ (no sum on } k). \end{aligned} \tag{48}$$

Since $b_{1,k}^{+}$ is sharply peaked at $\omega_k=\omega$, we make the approximations

$$\frac{b_{1,k}^{+}}{\omega_0 - \omega + \omega_k + \omega_{k'}} \approx \frac{b_{1,k}^{+}}{(\omega_0 + \omega_{k'})}$$
$$\frac{b_{1,k'}^{+}}{\omega_0 - \omega + \omega_k + \omega_{k'}} \approx \frac{b_{1,k'}^{+}}{(\omega_0 + \omega_k)} \tag{49}$$

with errors of order (36). With this approximation, the term involving $C_{k'}^2$ in Eq. (48) is equal to $i\Delta_{+}(\omega_0)b_{1,k}^{+}$. This cancels the $-i\Delta_{+}(\omega_0)b_{1,k}^{+}$ contribution from the second term on the right-hand side, giving

$$b_{1,k}^{+} = \frac{iC_k}{\omega_k - \omega - i\epsilon}\left[-b_{2,0}^{+} - \frac{\chi}{\omega_0 + \omega_k} - \frac{iC_{k'}b_{1,k'}^{+}}{\omega_0 - \omega + \omega_k + \omega_{k'}}\right]. \tag{50}$$

For future reference, we also note that

$$\frac{1}{(\omega_k - \omega - i\epsilon)}\frac{1}{(\omega_0 + \omega_k)} \approx \frac{1}{\Delta^{(+)}}\left[\frac{1}{\omega_k - \omega - i\epsilon} - \frac{1}{\omega_0 + \omega_k}\right]. \tag{51}$$

When the solution (50) is substituted into (42a), we obtain

$$b_{2,0}^{+} = \frac{1}{\tilde{\gamma} + i\Delta}\left[i\chi - \frac{\tilde{\gamma}\chi}{\Delta^{(+)}} - i\frac{C_{k'}C_k^2 b_{1,k'}^{+}}{(\omega_0 - \omega + \omega_k + \omega_{k'})(\omega_k - \omega - i\epsilon)}\right], \tag{52}$$

where

$$i\tilde{\gamma} = \lim_{\epsilon\to 0}\frac{\Delta^{(+)}C_k^2}{(\omega_k - \omega - i\epsilon)(\omega_0 + \omega_k)} = i\Gamma_{-}(\omega) + \Delta_{-}(\omega) - \Delta_{+}(\omega_0). \tag{53}$$

We have used

$$\lim_{\epsilon\to 0^{+}}\frac{1}{x - i\epsilon} = i\pi\delta(x) + P\left(\frac{1}{x}\right), \tag{54}$$

where P again denotes the Cauchy principal part.

Using the approximation

$$\begin{aligned} &\frac{b_{1,k'}^{+}}{(\omega_0 - \omega + \omega_k + \omega_{k'})(\omega_k - \omega - i\epsilon)} \\ &= \frac{b_{1,k'}^{+}}{\omega_{k'} + \omega_0}\left[\frac{1}{\omega_k - \omega - i\epsilon} - \frac{1}{\omega_0 - \omega + \omega_k + \omega_{k'}}\right] \\ &\approx \frac{b_{1,k'}^{+}}{\omega_{k'} + \omega_0}\left[\frac{1}{\omega_k - \omega - i\epsilon} - \frac{1}{\omega_0 + \omega_k}\right], \end{aligned} \tag{55}$$

we rewrite Eq. (52) as

$$b_{2,0}^{+} = \frac{1}{\tilde{\gamma} + i\Delta}\left[i\chi - \frac{\tilde{\gamma}\chi}{\Delta^{(+)}} - i\frac{\tilde{\gamma}C_k b_{1,k}^{+}}{\omega_k + \omega_0}\right]. \tag{56}$$

When Eq. (56), in turn, is substituted back into Eq. (50) and (49) is used, we finally obtain the following (integral) equation for $b_{1,k}^{+}$:

$$b_{1,k}^{+} = \frac{iC_k}{\omega_k - \omega - i\epsilon}\left\{\left(\frac{-i\chi}{\tilde{\gamma} + i\Delta} - \frac{\chi}{\omega_0 + \omega_k} + \frac{\tilde{\gamma}\chi}{\Delta^{(+)}(\tilde{\gamma} + i\Delta)}\right) - i\left(1 - \frac{\tilde{\gamma}}{(\tilde{\gamma} + i\Delta)}\right)\frac{C_{k'}b_{1,k'}^{+}}{\omega_0 + \omega_{k'}}\right\}. \tag{57}$$

This equation can be solved iteratively to evaluate the sums over k' that occur in each iteration. In doing so, we can make the approximation

$$\frac{C_k^2}{\omega_k-\omega-i\epsilon}\left(\frac{1}{\omega_0+\omega_k}\right)^2 = \frac{C_k^2}{\Delta^{(+)}}\left[\frac{1}{\omega_k-\omega-i\epsilon}-\frac{1}{\omega_0+\omega_k}\right]\frac{1}{\omega_0+\omega_k} \approx \frac{C_k^2}{\Delta^{(+)}}\frac{1}{\omega_k-\omega-i\epsilon}\frac{1}{\omega_0+\omega_k}=\frac{i\tilde{\gamma}}{\Delta^{(+)}},$$

the omitted term being of order (36). In effect, we can replace $\chi/(\omega_0+\omega_k)$ in (57) by $\chi/\Delta^{(+)}$ since this leads only to corrections of order (36) in calculating β. Carrying out the iteration using these approximations, we have

$$b_{1,k}^{+}=-\frac{i\chi C_k}{\omega_k-\omega-i\epsilon}(A+B)\sum_{n=0}^{\infty}[i\tilde{\gamma}B]^n, \tag{58}$$

where

$$A=\frac{i}{\tilde{\gamma}+i\Delta}$$

$$B=\frac{1}{\Delta^{(+)}}\left(1-\frac{\tilde{\gamma}}{(\tilde{\gamma}+i\Delta)}\right)=\frac{i\Delta}{\Delta^{(+)}(\tilde{\gamma}+i\Delta)}.$$

Since

$$|\tilde{\gamma}B|=\left|\frac{i\tilde{\gamma}\Delta}{\Delta^{(+)}(\tilde{\gamma}+i\Delta)}\right|\le\sqrt{\left|\frac{\tilde{\gamma}}{\Delta^{(+)}}\right|}\le\sqrt{\left|\frac{\tilde{\gamma}}{\omega_0}\right|}<1, \tag{59}$$

we can carry out the summation in Eq. (58) and obtain

$$b_{1,k}^{+}=\frac{-i\chi C_k}{\omega_k-\omega-i\epsilon}\frac{(A+B)}{1-i\tilde{\gamma}B}=\frac{-i\chi C_k}{\omega_k-\omega-i\epsilon}\frac{2\omega_0}{\Delta\Delta^{(+)}-2i\omega_0\tilde{\gamma}}, \tag{60}$$

where the identity $\Delta+\Delta^{(+)}=2\omega_0$ has been used. It then follows from Eqs. (52) and (60) that

$$b_{2,0}^{+}=\frac{-\chi}{\tilde{\gamma}+i\Delta}\left[-i+\frac{\tilde{\gamma}}{\Delta^{(+)}}+\frac{\tilde{\gamma}C_k^2}{(\omega_k-\omega-i\epsilon)(\omega_k+\omega_0)}\frac{2\omega_0}{\Delta\Delta^{(+)}-2i\omega_0\tilde{\gamma}}\right] =\frac{-\chi}{\tilde{\gamma}+i\Delta}\left[-i+\frac{\tilde{\gamma}}{\Delta^{(+)}}+\frac{i\tilde{\gamma}^2}{\Delta^{(+)}}\frac{2\omega_0}{\Delta\Delta^{(+)}-2i\omega_0\tilde{\gamma}}\right] =\frac{\chi\Delta^{(+)}}{\Delta\Delta^{(+)}-2i\omega_0\tilde{\gamma}}. \tag{61}$$

This completes the calculation of the "+" terms.

Although Eq. (46) is exact, we cannot apply it directly to obtain solutions for $b_{2,0}^{-}$, $b_{1,k}^{-}$, $b_{2,kk'}^{-}$ since we have assumed implicitly that $\omega>0$ in obtaining the solution (61). On the other hand, it is easy to carry out an iterative solution to Eqs. (42) to obtain, for instance,

$$b_{2,0}^{-}=\frac{\chi}{\Delta^{(+)}}. \tag{62}$$

All other amplitudes contribute to β amounts that are smaller by factors of order (36). In this manner we obtain, to this order, the combination $[\tilde{b}_{2,k}(b_{1,k}^{-})^*+(b_{2,0}^{-})^*]$ needed in the evaluation of β as

$$[\tilde{b}_{2,k}(b_{1,k}^{-})^*+(b_{2,0}^{-})^*]\approx\frac{\chi}{\Delta^{(+)}}. \tag{63}$$

Combining Eqs. (63), (39), (60), (61), and (45), we arrive at

$$\beta(\omega)=\frac{\Delta^{(+)}}{\Delta\Delta^{(+)}-2i\omega_0\tilde{\gamma}}+\left[\left(\frac{C_k^2}{(\omega_k-\omega-i\epsilon)(\omega_k+\omega_0)}\right)^*\frac{2\omega_0}{\Delta\Delta^{(+)}+2i\omega_0\tilde{\gamma}}\right]^*+\frac{1}{\Delta^{(+)}}=\frac{\Delta^{(+)}}{\Delta\Delta^{(+)}-2i\omega_0\tilde{\gamma}}+\frac{2i\omega_0\tilde{\gamma}/\Delta^{(+)}}{\Delta\Delta^{(+)}-2i\omega_0\tilde{\gamma}}+\frac{1}{\Delta^{(+)}} =\frac{2\omega_0}{\Delta\Delta^{(+)}-2i\omega_0\tilde{\gamma}} =\frac{2\omega_0}{\Delta\Delta^{(+)}+2\omega_0[\Delta_+(\omega_0)-\Delta_-(\omega)]-2i\omega_0\Gamma_-(\omega)}. \tag{64}$$

This expression is exact, subject to the conditions (36), and it is easily verified that it satisfies the optical theorem [Eq. (34)]. The polarizability calculated in this Schrödinger-picture approach is equivalent to that calculated in the Heisenberg picture when (36) holds and only contributions up to second order in d are retained. This can be seen by writing the factor in large brackets in (26) as $1+x\cong 1/(1-x)$, setting $\Gamma_+(\omega)=0$, and neglecting terms of order $[\Delta_+(\omega)-\Delta_+(\omega_0)]/\Delta$, consistent with condition (36b). This would imply that the condition $x\ll 1$, along with (36a), is necessary for the validity of our factorization hypotheses. We now turn again to the Heisenberg picture, using an approximation scheme that further supports this contention and helps to elucidate some aspects of the TLA polarizability.

IV. HEISENBERG PICTURE II

From the Hamiltonian (13) and the equal-time commutation relations, we obtain

$$\langle\dot{\sigma}\rangle=-i\omega_0\langle\sigma\rangle-2i\chi\cos\omega t\langle\sigma_z\rangle+C_k(\langle a_k\sigma_z\rangle-\langle\sigma_z a_k^\dagger\rangle), \tag{65}$$

$$\langle\dot{a}_k\rangle=-i\omega_k\langle a_k\rangle+C_k(\langle\sigma\rangle+\langle\sigma^\dagger\rangle). \tag{66}$$

Consistent with our previous approximations, we approximate $\langle\sigma_z\rangle$ in the second term on the right-hand side of (65) by -1, since we are neglecting any excited-state population in the effect of the applied field on the atom. Thus, we work with the approximate evolution equations

$$\langle\dot{\sigma}\rangle=-i\omega_0\langle\sigma\rangle+2i\chi\cos\omega t-C_k(\langle a_k\rangle-\langle a_k\rangle^*)+2C_k(\langle a_k\sigma_{22}\rangle-\langle a_k^\dagger\sigma_{22}\rangle), \tag{67a}$$

$$\langle \dot{a}_k \rangle = -i\omega_k \langle a_k \rangle + C_k(\langle \sigma \rangle + \langle \sigma^\dagger \rangle), \tag{67b}$$

$$\langle \sigma^\dagger \rangle = \langle \sigma \rangle^*, \tag{67c}$$

$$\langle a_k^\dagger \rangle = \langle a_k \rangle^*, \tag{67d}$$

where we have used $\sigma_z = \sigma_{22} - \sigma_{11} = 2\sigma_{22} - 1$.

It is now necessary to obtain equations of motion for $\langle a_k \sigma_{22} \rangle$ and its conjugate. This leads to an infinite set of coupled equations; however, since we are working to first order in χ, the equations can be truncated, just as in the amplitude approach. In fact, we can be guided by the amplitude approach in determining which operators must be retained. For example, $\langle a_k \sigma_{22} \rangle = \rho_{2k,2} = b_{2k} b_2^*$ and must be retained, whereas $\langle a_k^\dagger \sigma \rangle = \rho_{2,1k} = b_2 b_{1k}^*$ is of order χ^2 and can be neglected. In this way, we obtain a closed set of operator equations for the quantities

$$f_k = \langle a_k \sigma_{22} \rangle \tag{68a}$$

$$y_{kk'} = \langle a_{k'}^\dagger a_k \sigma \rangle \tag{68b}$$

$$z_{kk'} = \langle a_{k'} a_k \sigma \rangle \tag{68c}$$

$$c_k = \langle a_k \sigma \rangle \tag{68d}$$

$$x_{kk'k''} = \langle a_{k''}^\dagger a_{k'} a_k \sigma_{22} \rangle \tag{68e}$$

of the form

$$\frac{df_k}{dt} = -i\omega_k f_k + C_{k'} y_{k'k} + C_{k'} z_{kk'} + C_{k'} y_{kk'} - 2i\chi c_k \cos \omega t + C_k \langle \sigma^\dagger \rangle \tag{69a}$$

$$\frac{dy_{kk'}}{dt} = -i(\omega_0 + \omega_k - \omega_{k'}) y_{kk'} + C_{k''} x_{kk''k'} - C_{k''} x_{k'k''k}^* + C_{k'} f_k + C_k \langle a_{k'}^\dagger \rangle \tag{69b}$$

$$\frac{dz_{kk'}}{dt} = -i(\omega_0 + \omega_k + \omega_{k'}) z_{kk'} - C_k \langle a_{k'} \rangle + C_{k'} \langle a_k \rangle - C_{k''} x_{kk'k''} \tag{69c}$$

$$c_k = -i \frac{C_k}{\omega_0 + \omega_k}; \tag{69d}$$

$$\frac{dx_{kk'k''}}{dt} = -i(\omega_k + \omega_{k'} - \omega_{k''}) x_{kk'k''} - C_{k''} z_{kk'} + C_{k'} y_{kk''} - C_k y_{k'k''}, \tag{69e}$$

It can be shown that the contributions from the $x_{kk'k''}$, $z_{kk'}$, and $y_{kk''}$ terms are down by order of radiative shifts or widths divided by ω_0, and therefore can be neglected, consistent with condition (36a). Thus, the relevant equations are

$$\frac{df_k}{dt} = -i(\omega_k - i\epsilon) f_k - 2i\chi c_k \cos \omega t + C_k \langle \sigma^\dagger \rangle \tag{70a}$$

$$c_k = -\frac{C_k^*}{\omega_0 + \omega_k}, \tag{70b}$$

where the convergence parameter ϵ has been reintroduced.

The key point now is to assume trial solutions of the form

$$\langle \sigma \rangle = s e^{-i\omega t} + r e^{i\omega t} \tag{71a}$$

$$\langle \sigma^\dagger \rangle = r^* e^{-i\omega t} + s^* e^{i\omega t} \tag{71b}$$

$$\langle a_k \rangle = a_k^+ e^{-i\omega t} + a_k^- e^{i\omega t} \tag{71c}$$

$$f_k = f_k^+ e^{i\omega t} + f_k^- e^{-i\omega t}. \tag{71d}$$

In terms of these variables, the polarizability $\alpha(\omega)$ and $\beta(\omega)$, introduced in Secs. II and III, respectively, are given by

$$\alpha(\omega) = \frac{2d(s + r^*)}{E_0}; \quad \beta(\omega) = \frac{(s + r^*)}{\chi}. \tag{72}$$

Using the trial solution in the original equations, one finds steady-state equations

$$0 = -i\Delta s + i\chi - [C_k a_k^+ - C_k (a_k^-)^*] - [C_k f_k^+ - C_k (f_k^-)^*] \tag{73a}$$

$$0 = i\Delta^{(+)} r^* - i\chi + [C_k a_k^+ - C_k (a_k^-)^*] + [C_k f_k^+ - C_k (f_k^-)^*] \tag{73b}$$

$$0 = -i(\omega_k - \omega - i\epsilon) a_k^+ + C_k [s + r^*] \tag{73c}$$

$$0 = i(\omega_k + \omega)(a_k^-)^* + C_k [s + r^*] \tag{73d}$$

$$0 = -i(\omega_k - \omega - i\epsilon) f_k^+ - i\chi c_k + C_k r^* \tag{73e}$$

$$0 = i(\omega_k + \omega)(f_k^-)^* + i\chi c_k^* + C_k s \tag{73f}$$

$$c_k = -\frac{iC_k}{\omega_0 + \omega_k}. \tag{73g}$$

The solution of these equations is now straightforward, with

$$a_k^+ = \frac{-iC_k(s + r^*)}{\omega_k - \omega - i\epsilon} \tag{74a}$$

$$(a_k^-)^* = \frac{iC_k(s + r^*)}{\omega_k + \omega} \tag{74b}$$

and

$$-i[C_k f_k^+ - C_k (f_k^-)^*] = \chi g(\omega) - i\gamma_-(\omega) r^* - \Delta_+(\omega) s, \tag{75}$$

where

$$g(\omega) = i\frac{\gamma_-(\omega)}{\Delta^{(+)}} - \frac{\Delta_+(\omega_0)}{\Delta^{(+)}} - \frac{\Delta_+(\omega_0) - \Delta_+(\omega)}{\Delta} \tag{76}$$

and

$$\gamma_-(\omega) = \Gamma_-(\omega) - i\Delta_-(\omega).$$

When these results are substituted into Eqs. (73a) and (73b), one finds

$$0 = -i\Delta s + i\chi - g(\omega)(s + r^*) + 2i[\chi g(\omega) - i\gamma_-(\omega)r^* - \Delta_+(\omega)s], \quad (77)$$

$$0 = i\Delta^{(+)}r^* - i\chi + g(\omega)(s + r^*) - 2i[\chi g(\omega) - i\gamma_-(\omega)r^* - \Delta_+(\omega)s], \quad (78)$$

which can be solved to give

$$\beta = \frac{2\omega_0\left(1 + 2\frac{i\Gamma_-(\omega)}{\Delta^{(+)}} - 2\frac{\Delta_+(\omega_0) - \Delta_-(\omega)}{\Delta^{(+)}} - 2\frac{\Delta_+(\omega_0) - \Delta_+(\omega)}{\Delta}\right)}{\Delta\Delta^{(+)} - 2\omega[i\Gamma_-(\omega) + \Delta_-(\omega) - \Delta_+(\omega)]}. \quad (79)$$

Note that the last term in the numerator does not diverge as Δ goes to zero owing to the definition (8). Equation (79) coincides exactly with the polarizability (26) obtained in Sec. II, since $\Gamma_+(\omega)=0$ for $\omega>0$. Moreover, since all the fractions in the numerator are assumed to be small, we can move them to the denominator. In this way, using Eq. (43), one obtains

$$\beta = \frac{2\omega_0}{\Delta\Delta^{(+)} + 2\omega_0[-i\Gamma_-(\omega) - \Delta_-(\omega) - \Delta_+(\omega) + 2\Delta_+(\omega_0)]} \quad (80)$$

in agreement with the result of the amplitude approach (64) if one sets $[\Delta_+(\omega)-\Delta_+(\omega_0)]/\Delta \approx 0$, consistent with conditions (36b).

Thus, it appears that the validity conditions for the Heisenberg approach are that (36a) holds and that the ratios in the numerator of Eq. (79) are much less than unity. On the other hand, the Schrödinger approach is valid provided both conditions (36a) and (36b) are satisfied. It can be shown that Eq. (80) agrees exactly with a perturbative solution of Eqs. (38) carried out to second order in the vacuum coupling.

There are two features of Eq. (80) that merit some discussion. First, we see that the excited state Lamb shift, $-\hbar\Delta_-(\omega_0)$, that would enter the calculation is replaced by a "Lamb shift" evaluated at ω rather than ω_0. In fact, $\Delta_-(\omega)$ must be viewed as a level shift resulting from the *combined* dynamics of the applied and vacuum field. This result implies that Lamb shifts associated with excited states must be calculated within the context of a given problem. For example, in spontaneous emission from an atom prepared in the excited state, $\Delta_-(\omega_0)$ is the relevant shift parameter associated with the excited state [16]; however, in the case we consider here of an atom driven by an external field the relevant parameter is $\Delta_-(\omega)$. The frequency denominator in Eq. (80) contains the differences $[\Delta_-(\omega)-\Delta_+(\omega_0)]$ and $[\Delta_-(-\omega) - \Delta_+(\omega_0)]$, which can be interpreted as the differences between the excited state level shifts associated with the resonant and antiresonant components of the driving field, minus the ground state Lamb shift. The ground state Lamb shift is the only shift that is independent of the atom-field dynamics.

The second point to note is that the use of an "intuitive"-type expression for the polarizability resulting from the resonant and antiresonant components of the driving field, such as that implied in an expression given by Sakurai [2] (neglecting level shifts),

$$\beta_1 = \frac{1}{\Delta - i\Gamma_-(\omega)} + \frac{1}{\Delta^{(+)}} \quad (81)$$

leads to a result,

$$\frac{\text{Im}\,\beta_1}{\Gamma_-(\omega)|\beta_1|^2} \approx \left(\frac{2\omega_0}{\Delta^{(+)}}\right)^2,$$

which is consistent with the optical theorem $[\text{Im}[\beta] = \Gamma_-(\omega)|\beta|^2]$ only near resonance, when $2\omega_0/\Delta^{(+)} = \frac{2\omega_0}{\omega_0+\omega} \approx 1$. In contrast, our expression [Eq. (80)] (neglecting level shifts)

$$\beta = \frac{2\omega_0/\Delta^{(+)}}{\Delta - i(2\omega_0/\Delta_+)\Gamma_-(\omega)}$$

is consistent with the optical theorem for any atom-field detuning. Clearly, if one uses Eq. (33) $\{P=2\hbar\omega\chi^2\,\text{Im}[\beta]\}$, for the energy absorbed from the field, the use of Eq. (81) leads to considerable errors for large detunings. For example, if $\Delta/\omega_0=1/2$, $\Delta^{(+)}/\omega_0=3/2$, then

$$\text{Im}\,\beta_1 \approx \frac{\Gamma_-(\omega)}{\Delta^2}$$

while

$$\text{Im}\,\beta \approx \frac{\Gamma_-(\omega)}{\Delta^2}\left(\frac{2\omega_0}{\Delta^{(+)}}\right)^2 = \frac{9}{4}\frac{\Gamma_-(\omega)}{\Delta^2}.$$

V. CONCLUDING REMARKS

The expression (26) [or (80)] for the polarizability coincides, with one small difference, with that derived by Loudon and Barnett [12]. The difference is that the factor in large brackets in (26) is missing a term $-2C_k^2/(\omega_k+\omega_0)^2$ that appears in the Loudon-Barnett expression. In our approach, we have ignored terms of this order by consistently invoking (36a) [17].

As noted in the Introduction, a TLA that remains with high probability in its ground state is often approximated by a harmonic, Lorentzian oscillator obtained by replacing σ_z everywhere by -1. The polarizability for this model is then obtained using Eq. (14), replacing σ_z by -1, and following exactly the same approach as in Sec. II except that no "factorization" approximations, such as (15) or (18), are needed because the model is now linear in $\langle\sigma(t)\rangle$. One easily obtains

$$\alpha(\omega) = \frac{2d^2\omega_0}{\hbar}\frac{1}{\omega_0^2 - \omega^2 - 2i\omega_0[\gamma^{(-)}(\omega) - i\delta^{(+)}(\omega)]}, \quad (82)$$

with $\gamma^{(-)}(\omega)=\Gamma_-(\omega)-\Gamma_+(\omega)$ and $\delta^{(+)}(\omega)=\Delta_-(\omega)+\Delta_+(\omega)$. Although this expression satisfies the optical theorem, it has the unphysical feature that the radiative frequency shift $\delta^{(+)}(\omega)$ involves the *sum* of the radiative level shifts. The radiative frequency shift $\delta(\omega)$ in the polarizability (26), on the other hand, involves the expected *difference* in the two radiative level shifts. This point explains one of the reasons for including the radiative shifts in our calculations, even though, as noted earlier, these shifts cannot be properly renormalized in a two-level model to obtain physically meaningful Lamb shifts.

The familiar Lorentzian oscillator approximation to a TLA therefore fails to provide a physically satisfactory expression for the polarizability, even when the TLA has negligible excitation probability. The reason for this is clear from Eq. (14): for a TLA near its ground state, we can approximate $\langle\sigma_{22}(t)\rangle$ by 0 in the interaction of the atom with the applied field, but we cannot replace the *operator* $\sigma_{22}(t)$ by 0 when it appears in operator-product expectation values, such as $\langle a_k(t)\sigma_{22}(t)\rangle$. To first order in the external field amplitude, corrections to $\langle\sigma_{22}(t)\rangle=0$ are of order (36a) (the excited state population in the *absence* of any applied fields, resulting solely from the vacuum field), whereas contributions arising from $f_k=\langle a_k(t)\sigma_z(t)\rangle$ [Eq. (75)] are of order $\gamma(\omega)/\Delta^{(+)}(\omega)$ and must be included. As noted previously, $\langle a_k\sigma_{22}\rangle=\rho_{2k,2}=b_{2k}b_2^*$; this expectation value arises from interference between processes associated with the excitation of the atom by the external field and the antiresonant component of the vacuum field.

ACKNOWLEDGMENTS

We thank S. M. Barnett and R. Loudon for sharing their insights on this problem and for providing us with a preprint of their paper. We also thank G. W. Ford and J. E. Sipe for brief but helpful remarks relating to this work. This research is supported in part by the National Science Foundation through Grants No. PHY0244841 and No. ECS-0355206, and the FOCUS Grant, and by the US Army Research Office through a MURI grant, and by the Michigan Center for Theoretical Physics.

[1] H. A. Kramers and W. Heisenberg, Zeits. f. Physik. **31**, 681 (1925). An English translation of this paper, and a discussion of its importance in the development of quantum theory, may be found in B. L. van der Waerden, *Sources of Quantum Mechanics* (Dover, New York, 1968).

[2] See, for instance, J. J. Sakurai, *Advanced Quantum Mechanics* (Addison-Wesley, Reading, MA, 1976), Eq. (2.188). Sakurai notes that the optical theorem is exact, but that his expression for the scattering amplitude is perturbative and satisfies the optical theorem only in the vicinity of resonance.

[3] See, for instance, D. L. Andrews, S. Naguleswaran, and G. E. Stedman, Phys. Rev. A **57**, 4925 (1998); G. E. Stedman, S. Naguleswaran, D. L. Andrews, and L. C. Dávila Romero, *ibid.* **63**, 047801 (2001); D. L. Andrews, L. C. Dávila Romero, and G. E. Stedman, *ibid.* **67**, 055801 (2003).

[4] See for instance, A. D. Buckingham and P. Fischer, Phys. Rev. A **61**, 035801 (2000); Phys. Rev. A **63**, 047802 (c4 c4a c4b c4cR. W. Boyd, *Nonlinear Optics*, 2nd ed. (Academic Press, New York, 158.

[5] The relevance of the damping terms to the question of the existence of a linear electro-optic effect in isotropic chiral media is discussed by R. W. Boyd, J. E. Sipe, and P. W. Milonni, J. Opt. A, Pure Appl. Opt. **6**, S14 (2004).

[6] G. S. Agarwal and R. W. Boyd, Phys. Rev. A **67**, 043821 (2003).

[7] D. A. Long, *The Raman Effect* (Wiley, New York, 2001).

[8] See, for instance, J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975), p. 423.

[9] P. W. Milonni and R. W. Boyd, Phys. Rev. A **69**, 023814 (2004). There is a typographical error in Eq. (25): V in that equation should be replaced by $-V$.

[10] See, for instance, P. W. Milonni, *The Quantum Vacuum: An Introduction to Quantum Electrodynamics* (Academic, San Diego, 1994).

[11] Equation (12) follows directly from Eqs. (25) and (26) of MB when the approximation noted after Eq. (26) is not made.

[12] R. Loudon and S. M. Barnett, J. Phys. B **39**, S555 (2006).

[13] See, for example, P. R. Berman and B. Dubetsky, Phys. Rev. A **55**, 4060 (1997), and references therein.

[14] The level shifts do not include light shifts, proportional to the intensity of the external field, which are neglected in this work.

[15] The upper- and lower-state energy levels are now taken to be $\hbar\omega_0$ and 0, respectively, rather than $\pm\frac{1}{2}\hbar\omega_0$ as implied by the Hamiltonian (13).

[16] G. S. Agarwal, Phys. Rev. A **7**, 1195 (1973).

[17] It might be worth noting, however, that we can obtain a result in exact agreement with Loudon and Barnett Ref. [12] [an extra term equal to $-2C_k^2/(\omega_k+\omega_0)^2$ in the bracketed factor of (26)], if we turn on the vacuum field adiabatically and keep terms of this order. In effect, this extra term would correspond to the fact that there is a population difference between the "bare" ground and excited states when these states are dressed by the vacuum field. By neglecting terms of this order, we avoid corrections related to nonvanishing excited state populations in the absence of any external field.