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Theory of coherent two-photon resonance

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A theory of coherent two-photon resonance is developed by applying a canonical transformation on the Heisenberg equation of atomic motion. The theory leads both to the optical Bloch equation and the second-order induced polarization which drives the classical Maxwell's equation. The expressions for the Rabi frequency and the induced polarization are given explicitly for two-photon absorption and Raman effect. The results are applied to coherent steady-state propagation, and two kinds of pulses are derived for different-frequency two-photon absorption, including the atomic frequency shift.

I. INTRODUCTION

Recent progress in the study of coherent resonant interaction between light and a collection of atoms has led to the observations of such novel effects as self-induced transparency, photon echo, superradiance, optical nutation, optical free induction decay, and optical adiabatic inversion. Experimental and theoretical works have been reviewed in recent books.[1] However, these works have been limited almost exclusively to one-photon-resonance cases. Belenov and Poluektov[2] first discussed the coherent two-photon absorption, but a related experiment appeared only recently.[3] More recently, a new experiment has been reported in a resonant three-level system.[4]

In a previous paper,[5] the author examined the validity of the two-level approximation for atoms in the coherent resonant interaction and showed that other nonresonant energy levels yield Stark-like secular shifts of two energy levels under consideration, which may be important for two-photon resonance. In this paper, we shall discuss coherent two-photon two-level resonance using the same canonical transformation of Ref. 5. The main improvements over the previous paper are the following three points.

First, the Heisenberg equation is used for the description of atoms instead of the Schrödinger equation. For two-level-resonance phenomena, the Heisenberg method is much more straightforward. Secondly, the second-order induced polarizations which drive Maxwell's equation are given explicitly for two-photon absorption and the Raman effect. Lastly, the results are applied to coherent two-photon-resonance propagation effect.

II. CANONICAL TRANSFORMATION

The time-development of any operator A which is time dependent only implicitly may be described by the Heisenberg equation

$$i\hbar \frac{dA}{dt} = -[\mathcal{H}, A], \tag{1}$$

where $\mathcal{H}$ is the one-atom Hamiltonian which consists of the unperturbed part and the perturbation

$$\mathcal{H} = H - \vec{d} \cdot \vec{E}. \tag{2}$$

Here $\vec{d}$ is the electric dipole-moment operator and $\vec{E}$ the classical radiation field,

$$\vec{E} = 2\vec{\mathcal{E}}\, \epsilon \cos\varphi, \quad \varphi = \omega t - kz + \phi. \tag{3}$$

The unit polarization vector $\vec{\mathcal{E}}$, amplitude ϵ, phases φ and ϕ, and wave vector k are introduced in the usual way.

Although we are interested in the case where the field frequency ω is almost resonant with an atomic transition frequency ω_0, the real atom has many energy levels in addition to the resonant two levels assumed nondegenerate. Since two-photon resonance occurs inevitably through virtual nonresonant states, they must be taken into account from the onset. The perturbation Hamiltonian therefore gives rise to both resonant and nonresonant second-order processes. This situation is inconvenient for treating resonance phenomena. Thus we search for some canonical transformation which could eliminate the nonresonant interaction from the (revised) perturbation Hamiltonian and renormalize it into the unperturbed Hamiltonian. Then the Pauli-spin-operator formalism for the "two-level" atom would be applied straightforwardly to the revised Hamiltonian.

The canonical transformation in radiation theory is well known for quantum-electrodynamic self-energy renormalization. In our case the self-energy-type Stark shift is the nonresonant process which should be renormalized to the unperturbed atomic energy. After renormalization, only resonant processes have to be considered. We borrow the physical spirit from Heitler,[6] but the calculation is much simpler than quantum-electrodynamic theory because we treat the field classically. Compared with usual time-dependent perturbation theory, our approach has the advantage of systematic treatment of resonance phenomena of any kind and order.

The canonical transformation on A may be written as

$$\tilde{A}=e^{-iS}Ae^{iS}, \tag{4}$$

where S is a Hermitian operator. It is easy to show that $\tilde{A}$ obeys the equation

$$i\hbar\frac{d\tilde{A}}{dt}=-[K,\tilde{A}], \tag{5}$$

$$K=e^{-iS}\mathcal{H}e^{iS}+\hbar\frac{dS}{dt}. \tag{6}$$

We now determine the explicit matrix form of S so that the revised Hamiltonian K is divided definitely into the resonant and nonresonant parts. According to the above consideration, we rewrite the original Hamiltonian as

$$\mathcal{H}=\mathcal{H}^{(0)}+\mathcal{H}^{(1)}+\mathcal{H}^{(2)}, \tag{7a}$$

$$\mathcal{H}^{(0)}=H+H_S, \tag{7b}$$

$$\mathcal{H}^{(1)}=-\vec{\mathrm{d}}\cdot\vec{\mathrm{E}}, \tag{7c}$$

$$\mathcal{H}^{(2)}=-H_S, \tag{7d}$$

where H_S is the energy-shift operator. Since we are interested in two-photon effects, it is sufficient to take H_S to second order in $\vec{\mathrm{E}}$. Now $\mathcal{H}^{(0)}$ is regarded as the new zeroth-order Hamiltonian.

In terms of this new order, operators S and K are expanded,

$$S=S^{(1)}+S^{(2)}+\cdots, \tag{8a}$$

$$K=K^{(0)}+K^{(1)}+K^{(2)}+\cdots. \tag{8b}$$

Using the expansion

$$e^{-iS}\mathcal{H}e^{iS}=\mathcal{H}-i[S,\mathcal{H}]-\tfrac{1}{2}[S,[S,\mathcal{H}]]+\cdots, \tag{9}$$

substituting Eqs. (7) and (8) into (6), and equating the terms of the same order, we have the relations

$$K^{(0)}=\mathcal{H}^{(0)}, \tag{10a}$$

$$K^{(1)}=\mathcal{H}^{(1)}-i[S^{(1)},\mathcal{H}^{(0)}]+\hbar\, dS^{(1)}/dt, \tag{10b}$$

$$K^{(2)}=\mathcal{H}^{(2)}-i[S^{(1)},\mathcal{H}^{(1)}]-i[S^{(2)},\mathcal{H}^{(0)}]-\tfrac{1}{2}[S^{(1)},[S^{(1)},\mathcal{H}^{(0)}]]+\hbar\, dS^{(2)}/dt. \tag{10c}$$

The eigenstate of $K^{(0)}$ is chosen as the base,

$$K^{(0)}|m\rangle=\hbar\omega_m|m\rangle, \tag{11}$$

which is also the eigenstate of the original unperturbed Hamiltonian.[5] The eigenvalue $\hbar\omega_m$ may be time dependent when the system is subjected to optical pulses.

We choose $S^{(1)}$ and $S^{(2)}$ so that the new perturbation Hamiltonians $K^{(1)}$ and $K^{(2)}$ contain only terms connecting the two resonant states $|+\rangle$ and $|-\rangle$. The degree of resonance condition depends on the problem. We here require only vaguely that the off-resonance $\Delta=\omega_0-\omega$ must be small enough compared with the transition frequency nearest to ω_0, and denote the critical off-resonance by $\delta\omega$.

The requirement

$$K^{(1)}_{mn}=0,\quad ||\omega_{mn}|-\omega|>\delta\omega \tag{12}$$

on Eq. (10b) and the assumption that the eigenvalue variation is negligibly small compared with optical energy yield the result

$$S^{(1)}_{mn}=\frac{id_{mn}\epsilon}{\hbar}\left(\frac{e^{-i\varphi}}{\omega_{nm}+\omega}+\frac{e^{i\varphi}}{\omega_{nm}-\omega}\right), \tag{13}$$

where $d=\vec{\mathrm{d}}\cdot\vec{\mathrm{E}}$ and $\omega_{nm}=\omega_n-\omega_m$. For resonance, we require

$$S^{(1)}_{mn}=0,\quad ||\omega_{mn}|-\omega|<\delta\omega \tag{14}$$

in order to leave only the resonant term in $K^{(1)}$,

$$K^{(1)}_{+-}=-2d_{+-}\epsilon\cos\varphi. \tag{15}$$

Next in $K^{(2)}$, $-\mathcal{H}^{(2)}$ is chosen to be the Stark shift of the unperturbed energy,

$$(H_S)_{mm}=-\mathcal{H}^{(2)}_{mm}=-\tfrac{1}{2}i\langle[S^{(1)},\mathcal{H}^{(1)}]_{mm}\rangle_{\text{time av}}=-\sum_{l}\frac{2\omega_{lm}d^2_{lm}\epsilon^2}{\hbar(\omega^2_{lm}-\omega^2)}. \tag{16}$$

The requirement

$$K^{(2)}_{mn}=0,\quad ||\omega_{mn}|-\omega_c|>\delta\omega \tag{17}$$

on Eq. (10c) implies that

$$\hbar\frac{dS^{(2)}}{dt}-i[S^{(2)},\mathcal{H}^{(0)}]-\tfrac{1}{2}i[S^{(1)},\mathcal{H}^{(1)}]_{\text{opt}}=0, \tag{18}$$

where ω_c is a two-photon combination frequency and $[\ \ ]_{\text{opt}}$ means that only the optical-frequency term should be taken in the bracket. In deriving Eq. (18) we used the relations $K^{(1)}=0$, $[S^{(1)},dS^{(1)}/dt]=0$, and Eq. (16). For resonance, we require

$$S^{(2)}_{mn}=0,\quad ||\omega_{mn}|-\omega_c|<\delta\omega \tag{19}$$

in order to leave only the resonant term in $K^{(2)}$,

$$K^{(2)}_{+-}=-\tfrac{1}{2}i[S^{(1)},\mathcal{H}^{(1)}]_{+-,\text{opt}}. \tag{20}$$

From Eq. (20), expressions for the new interaction Hamiltonian $K^{(2)}_{+-}$ are explicitly calculated and given below for typical three cases. Two kinds of field are defined by

$$\vec{\mathrm{E}}_{L,s}=2\vec{\mathcal{E}}\epsilon_{L,s}\cos\varphi_{L,s},\quad \varphi_{L,s}=\omega_{L,s}t-k_{L,s}z+\phi_{L,s}.$$

(a) Two-photon absorption, $\omega_0\approx2\omega_L$,

$$K^{(2)}_{+-}=-[R^{+}_{L}e^{-2i\varphi_L}+R^{-}_{L}e^{2i\varphi_L}]\epsilon^2_L. \tag{21}$$

(b) Two-photon absorption, $\omega_0 \approx \omega_L + \omega_S$, $R_L^+ = R_S^+$,

$$K_{+-}^{(2)} = -[(R_L^+ + R_S^+)e^{-i(\varphi_L+\varphi_S)} + (R_L^- + R_S^-)e^{i(\varphi_L+\varphi_S)}]\epsilon_L\epsilon_S. \tag{22}$$

(c) Raman effect, $\omega_0 \approx \omega_L - \omega_S$, $R_L^+ = R_S^-$,

$$K_{+-}^{(2)} = -[(R_L^+ + R_S^-)e^{-i(\varphi_L-\varphi_S)} + (R_L^- + R_S^+)e^{i(\varphi_L-\varphi_S)}]\epsilon_L\epsilon_S. \tag{23}$$

Here

$$R_{L,S}^{\pm} = \sum_l \frac{d_{-l}d_{l+}}{2\hbar}\left(\frac{1}{\omega_{l+} \pm \omega_{L,S}} + \frac{1}{\omega_{l-} \mp \omega_{L,S}}\right). \tag{24}$$

In summary, the revised Hamiltonian is divided into the unperturbed Hamiltonian including the energy shift and the resonant perturbation Hamiltonian, which is written for two-photon resonance as

$$K = \mathcal{H}^{(0)} + K^{(2)}, \tag{25}$$

where $K^{(2)}$ is given by Eqs. (21)–(23) according to the problem of interest. Since $K^{(2)}$ is in general imaginary, we put

$$K^{(2)} = K_R^{(2)}\cos\varphi_c - iK_I^{(2)}\sin\varphi_c,$$

$$\varphi_c = 2\varphi_L \quad \text{or} \quad \varphi_L \pm \varphi_S. \tag{26}$$

III. OPTICAL BLOCH EQUATION

Since the perturbation Hamiltonian now connects only two resonant levels, it is sufficient to take the Hamiltonian in the form

$$K = \tfrac{1}{2}\hbar\omega_{+-}\sigma_3 + K_R^{(2)}\sigma_1\cos\varphi_c + K_I^{(2)}\sigma_2\sin\varphi_c, \tag{27}$$

where σ_i ($i = 1, 2, 3$) is the Pauli spin operator and the new transition frequency ω_{+-} is written using Eq. (16)

$$\omega_{+-} = \omega_0 + \alpha_{L,S}^{-}\epsilon_{L,S}^2, \tag{28a}$$

$$\alpha_{L,S}^{\mp} = \sum_{l \neq +,-}\left(\frac{2d_{-l}^2}{\hbar^2}\,\frac{\omega_{l-}}{\omega_{l-}^2 - \omega_{L,S}^2} \mp \frac{2d_{+l}^2}{\hbar^2}\,\frac{\omega_{l+}}{\omega_{l+}^2 - \omega_{L,S}^2}\right). \tag{28b}$$

A comparison of Eqs. (28b) and (24) shows that $\hbar\alpha_{L,S}^{-}$ is comparable with $R_L^{\pm}$ and therefore cannot be neglected for two-photon resonance.

Using the commutation relation

$$\vec{\sigma} \times \vec{\sigma} = 2i\vec{\sigma}, \quad \vec{\sigma} = (\sigma_1, \sigma_2, \sigma_3), \tag{29}$$

we can derive immediately the Heisenberg equations for spin operators,

$$\frac{d\sigma_1}{dt} = -\omega_{+-}\sigma_2 + \frac{2}{\hbar}K_I^{(2)}\sigma_3\sin\varphi_c, \tag{30a}$$

$$\frac{d\sigma_2}{dt} = \omega_{+-}\sigma_1 - \frac{2}{\hbar}K_R^{(2)}\sigma_3\cos\varphi_c, \tag{30b}$$

$$\frac{d\sigma_3}{dt} = \frac{2}{\hbar}K_R^{(2)}\sigma_2\cos\varphi_c - \frac{2}{\hbar}K_I^{(2)}\sigma_1\sin\varphi_c. \tag{30c}$$

Taking the expectations $s_i = \langle\sigma_i\rangle$, transforming s_i to the Bloch-vector variables

$$s_1 = u\cos\varphi_c - v\sin\varphi_c, \tag{31a}$$

$$s_2 = u\sin\varphi_c + v\cos\varphi_c, \tag{31b}$$

$$s_3 = w, \tag{31c}$$

and using the rotating-wave approximation, we have finally

$$du/dt = -\Omega_0 v, \tag{32a}$$

$$dv/dt = \Omega_0 u + \Omega w, \tag{32b}$$

$$w = -\Omega v, \tag{32c}$$

where

$$\Omega_0 = \omega_{+-} - \omega_c - \dot{\phi}_c = \Delta + \alpha_L^- \epsilon_L^2 + \alpha_S^- \epsilon_S^2 - \dot{\phi}_c, \tag{32d}$$

$$\Omega = -(K_R^{(2)} + K_I^{(2)})/2\hbar. \tag{32e}$$

The optical Bloch equation (32) is the analog of the usual one-photon-resonance equation. Essential differences are that Ω is now second order in the field amplitude and ω_{+-} in Ω_0 includes the atomic frequency shift. Using Eqs. (32e), (26), (25), and (21)–(23), we can obtain the expressions for Ω explicitly for the following two cases:

(a) Two-photon absorption, $\omega_0 \approx 2\omega_L$,

$$\Omega = 2R_L^+\epsilon_L^2/\hbar. \tag{33a}$$

(b) Two-photon absorption, $\omega_0 \approx \omega_L + \omega_S$, and Raman effect, $\omega_0 \approx \omega_L - \omega_S$,

$$\Omega = 4R_L^+\epsilon_L\epsilon_S/\hbar. \tag{33b}$$

IV. INDUCED POLARIZATION

The optical Bloch equation which describes the atomic motion with two-photon resonance has been derived in Sec. III. In this section we give the expressions for the atomic two-photon polarization which drives Maxwell's equation

$$\frac{\partial^2 \vec{\mathrm{E}}}{\partial z^2} - \frac{\eta^2}{c^2}\frac{\partial^2 \vec{\mathrm{E}}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 \vec{\mathrm{P}}}{\partial t^2}, \tag{34}$$

where η is the refractive index of the host medium. The induced polarization may be written in the usual manner as

$$P = \vec{\mathscr{E}} \cdot \vec{\mathrm{P}} = N[\mathrm{Tr}(\rho d)]_{\mathrm{av}}, \tag{35}$$

where N is the atomic density, ρ the density matrix, and $[\ ]_{\mathrm{av}}$ represents the average over the atomic resonance-frequency distribution. Using the transformed operators introduced in Sec. II,

P can be rewritten

$$P = N[\mathrm{Tr}(\tilde{\rho}\tilde{d})]_{\mathrm{av}}. \tag{36}$$

We now calculate explicitly the induced polarization (36) for two-photon resonance using the result of Sec. II.

The two-level density matrix $\tilde{\rho}$ can be written using the Pauli spin operator as[7]

$$\tilde{\rho} = \tfrac{1}{2}\,\mathrm{Tr}\tilde{\rho} + \tfrac{1}{2}\langle\vec{\sigma}\rangle\cdot\vec{\sigma} = \tfrac{1}{2} + \tfrac{1}{2}\vec{\mathrm{s}}\cdot\vec{\sigma}, \tag{37}$$

where $\vec{\mathrm{s}} = \langle\vec{\sigma}\rangle$ and the relation $\mathrm{Tr}\tilde{\rho} = 1$ is used. In a similar way, the operator $\tilde{d}$ is written

$$\tilde{d} = \tfrac{1}{2}\,\mathrm{Tr}\tilde{d} + \tfrac{1}{2}\vec{\mathrm{D}}\cdot\vec{\sigma}, \tag{38}$$

where

$$\vec{\mathrm{D}} = (2\tilde{d}_{+-,R}\,,\,-2\tilde{d}_{+-,I}\,,\,\tilde{d}_{++} - \tilde{d}_{--}) \tag{39}$$

and the subscripts R and I mean the real and imaginary parts. Using the operator identity

$$\sigma_k\sigma_i + \sigma_i\sigma_k = 2\delta_{ik}\,, \tag{40}$$

it is easy to prove the relation

$$P = N[\tfrac{1}{2}\,\mathrm{Tr}\tilde{d} + \tfrac{1}{2}\vec{\mathrm{s}}\cdot\vec{\mathrm{D}}]_{\mathrm{av}}. \tag{41}$$

Since we are interested in two-photon resonance, it is sufficient to take

$$\tilde{d} = e^{-iS}de^{iS} = d - i[S^{(1)}, d]. \tag{42}$$

The first term on the right-hand side contributes to the usual one-photon-resonance polarization

$$P^{(1)} = Nd_{+-}[s_1]_{\mathrm{av}} = Nd_{+-}[u\cos\varphi - v\sin\varphi]_{\mathrm{av}}\,. \tag{43}$$

The matrix elements of the second term, $-i[S^{(1)},d]$, can be obtained in a similar way with the calculations of Eq. (16) for $(H_S)_{mm}$ and Eq. (20) for $K^{(2)}_{+-}$. For example, the first term in the brackets in Eq. (41) is calculated as

$$\tfrac{1}{2}\,\mathrm{Tr}\tilde{d} = -\tfrac{1}{2}i\,\mathrm{Tr}[S^{(1)}, d] = \hbar\alpha^{+}_{L,S}\epsilon_{L,S}\cos\varphi_{L,S} \tag{44}$$

where $\alpha^{+}_{L,S}$ is given in Eq. (28b). The calculation of the second term in Eq. (41) is straightforward but rather involved. We therefore give only the final results, after transforming s_i to the Bloch-vector variables according to Eq. (31), for the three cases

(a) Two-photon absorption, $\omega_0 \approx 2\omega_L$,

$$P^{(2)}_L = N[(R^{+}_L u - \tfrac{1}{2}\hbar\,\alpha^{-}_L w + \tfrac{1}{2}\hbar\,\alpha^{+}_L)2\epsilon_L\cos\varphi_L - (R^{+}_L v)2\epsilon_L\sin\varphi_L]_{\mathrm{av}}\,. \tag{45a}$$

(b) Two-photon absorption, $\omega_0 \approx \omega_L + \omega_S$, $R^{+}_L = R^{+}_S$,

$$P^{(2)}_L = N[(R^{+}_L\epsilon_S u - \tfrac{1}{2}\hbar\alpha^{-}_L\epsilon_L w + \tfrac{1}{2}\hbar\alpha^{+}_L\epsilon_L)2\cos\varphi_L - (R^{+}_L\epsilon_S v)2\sin\varphi_L]_{\mathrm{av}}\,, \tag{45b}$$

$$P^{(2)}_S = N[(R^{+}_L\epsilon_L u - \tfrac{1}{2}\hbar\alpha^{-}_S\epsilon_S w + \tfrac{1}{2}\hbar\alpha^{+}_S\epsilon_S)2\cos\varphi_S - (R^{+}_L\epsilon_L v)2\sin\varphi_S]_{\mathrm{av}}\,. \tag{45c}$$

(c) Raman effect, $\omega_0 \approx \omega_L - \omega_S$, $R^{+}_L = R^{-}_S$,

$$P^{(2)}_L = N[(R^{+}_L\epsilon_S u - \tfrac{1}{2}\hbar\alpha^{-}_L\epsilon_L w + \tfrac{1}{2}\hbar\alpha^{+}_L\epsilon_L)2\cos\varphi_L - (R^{+}_L\epsilon_S v)2\sin\varphi_L]_{\mathrm{av}}\,, \tag{45d}$$

$$P^{(2)}_S = N[(R^{+}_L\epsilon_L u - \tfrac{1}{2}\hbar\alpha^{-}_S\epsilon_S w + \tfrac{1}{2}\hbar\alpha^{+}_S\epsilon_S)2\cos\varphi_S + (R^{+}_L\epsilon_L v)2\sin\varphi_S]_{\mathrm{av}}\,. \tag{45e}$$

These induced polarizations are coupled with Maxwell's equation (34). When P is written as the sum of the in-phase and in-quadrature components as

$$P^{(2)} = N(U\cos\varphi - V\sin\varphi), \tag{46}$$

the slowly-varying Maxwell's equations yield

$$\frac{\partial\epsilon}{\partial z} + \frac{\eta}{c}\,\dot{\epsilon} = \frac{N\pi\omega}{c\eta}\,V, \tag{47a}$$

$$\frac{\partial\phi}{\partial z} + \frac{\eta}{c}\,\dot{\phi} = \frac{N\pi\omega}{c\eta\epsilon}\,U. \tag{47b}$$

The optical Bloch equation (32) and Maxwell's equation (47), with the induced polarization (45) and (46), are the fundamentals for treating coherent two-photon-resonance propagation. Sec. V deals with this propagation effect in steady state.

V. PULSE PROPAGATION IN STEADY STATE

The combined optical Bloch and Maxwell's equations are very complicated, especially for two-photon resonance. Even for one-photon resonance, analytic solutions are possible only for some special cases such as steady-state propagation.[1] We show below that the steady-state *pulse* propagation for two-photon absorption allows an analytic solution. Only the sharp-line case is discussed for simplicity.

Comparing Eqs. (45) and (46b)–(46e), we have from Eqs. (47) the field equations

$$\frac{\partial\epsilon_L}{\partial z} + \frac{\eta_L}{c}\,\dot{\epsilon}_L = \frac{2\pi\omega_L N}{c\eta_L}\,R^{+}_L\epsilon_S v, \tag{48a}$$

$$\frac{\partial\epsilon_S}{\partial z} + \frac{\eta_S}{c}\,\dot{\epsilon}_S = \pm\frac{2\pi\omega_S N}{c\eta_S}\,R^{+}_L\epsilon_L v, \tag{48b}$$

$$\frac{\partial\phi_L}{\partial z} + \frac{\eta_L}{c}\,\dot{\phi}_L = -\frac{2\pi\omega_L N}{c\eta_L\epsilon_L}\,(R^{+}_L\epsilon_S u - \tfrac{1}{2}\hbar\alpha^{-}_L\epsilon_L w + \tfrac{1}{2}\hbar\alpha^{+}_L\epsilon_L), \tag{48c}$$

$$\frac{\partial\phi_S}{\partial z} + \frac{\eta_S}{c}\,\dot{\phi}_S = -\frac{2\pi\omega_S N}{c\eta_S\epsilon_S}\,(R^{+}_L\epsilon_L u - \tfrac{1}{2}\hbar\alpha^{-}_S\epsilon_S w + \tfrac{1}{2}\hbar\alpha^{+}_S\epsilon_S). \tag{48d}$$

In Eq. (48b), + and − correspond to two-photon absorption and Raman effect, respectively. Steady-state equations are given as usual by the transformation, $\partial\epsilon/\partial z \to -\dot{\epsilon}/V_0$, where the differentiation is with respect to the local time,

$$\zeta = t - z/V_0, \tag{49}$$

and V_0 is the constant velocity. Introducing constants

$$Q_{L,S} = 2\pi\omega_{L,S} N R_L^+ V_0/\eta_{L,S}(c - \eta_{L,S} V_0), \tag{50}$$

we have from Eq. (48)

$$\dot{\epsilon}_L = -Q_L \epsilon_S v, \tag{51a}$$

$$\dot{\epsilon}_S = \mp Q_S \epsilon_L v, \tag{51b}$$

$$\dot{\phi}_L = Q_L\left(\frac{\epsilon_S}{\epsilon_L} u - \frac{\hbar\alpha_L^-}{2R_L^+} w + \frac{\hbar\alpha_L^+}{2R_L^+}\right), \tag{51c}$$

$$\dot{\phi}_S = Q_S\left(\frac{\epsilon_L}{\epsilon_S} u - \frac{\hbar\alpha_S^-}{2R_L^+} w + \frac{\hbar\alpha_S^+}{2R_L^+}\right). \tag{51d}$$

Equations (51a) and (51b) immediately lead to the relation

$$Q_S \epsilon_L^2 \mp Q_L \epsilon_S^2 = Q_S \epsilon_{L0}^2 \mp Q_L \epsilon_{S0}^2, \tag{52}$$

where ϵ_{L0} and ϵ_{S0} are the initial values and must be zero for single pulses. We hereafter confine our consideration to the experimentally most interesting case of *pulse* propagation. It is clearly seen from Eq. (52) that the steady-state single-pulse solution does not exist for the Raman effect. For two-photon absorption

$$Q_S \epsilon_L^2 = Q_L \epsilon_S^2. \tag{53}$$

With the relation (53), Maxwell's equations (51) reduce to simpler expressions

$$\dot{\epsilon}_L = -\sqrt{Q_L Q_S}\, \epsilon_L v, \tag{54a}$$

$$\dot{\phi}_L = \sqrt{Q_L Q_S}\, u - \frac{\hbar\alpha_L^- Q_L}{2R_L^+} w + \frac{\hbar\alpha_L^+ Q_L}{2R_L^+}, \tag{54b}$$

$$\dot{\phi}_S = \sqrt{Q_L Q_S}\, u - \frac{\hbar\alpha_S^- Q_S}{2R_L^+} w + \frac{\hbar\alpha_S^+ Q_S}{2R_L^+}, \tag{54c}$$

as well as the components Ω_0 [Eq. (32d)] and Ω [Eq. (33b)] of the torque vector

$$\Omega_0 = \Delta - \dot{\phi} + B\epsilon_L^2, \tag{55a}$$

$$\Omega = C\epsilon_L^2, \tag{55b}$$

where

$$B = \alpha_L^- + \alpha_S^- Q_S/Q_L, \quad C = (4R_L^+/\hbar)\sqrt{Q_S/Q_L}\,. \tag{55c}$$

Adding Eqs. (54b) and (54c), differentiating and inserting the expressions (32a) and (32c) for $\dot{u}$ and $\dot{w}$ with (55), we have

$$\ddot{\phi} = \ddot{\phi}_L + \ddot{\phi}_S = -2\sqrt{Q_L Q_S}\,(\Delta - \dot{\phi})v. \tag{56}$$

Comparison of Eqs. (56) and (54a) gives

$$\ddot{\phi}\epsilon_L = 2(\Delta - \dot{\phi})\dot{\epsilon}_L, \tag{57}$$

which means the relation

$$\epsilon_L^2(\dot{\phi} - \Delta) = \text{const.} \tag{58}$$

For a single pulse we have clearly

$$\dot{\phi} = \Delta. \tag{59}$$

That is, the phase modulation exactly cancels the off resonance in steady state. Then the Bloch equations (32) become the more transparent forms

$$\dot{u} = -B\epsilon_L^2 v, \tag{60a}$$

$$\dot{v} = +B\epsilon_L^2 u + C\epsilon_L^2 w, \tag{60b}$$

$$\dot{w} = -C\epsilon_L^2 v. \tag{60c}$$

It is easy to derive the differential equation for ϵ_L from Eqs. (54a) and (60a) to (60c). The result is

$$(\dot{\epsilon}_L^2) = \epsilon_L^2(C_1 + C_2\epsilon_L^2 - C_3\epsilon_L^4)^{1/2}, \tag{61a}$$

where

$$C_1 = 4Q_L Q_S v_0^2, \tag{61b}$$

$$C_2 = -4\sqrt{Q_L Q_S}\,(Bu_0 + Cw_0), \tag{61c}$$

$$C_3 = B^2 + C^2, \tag{61d}$$

and u_0, v_0, w_0 are the initial values for u, v, w. Equation (61a) has two types of solution, (a) $v_0 \neq 0$,

$$\epsilon_L^2 = \frac{4C_1 \exp[-\sqrt{C_1}\,(\zeta - \zeta_0)]}{\{\exp[-\sqrt{C_1}\,(\zeta - \zeta_0)] - C_2\}^2 + 4C_1C_3}, \tag{62a}$$

where ζ_0 is the integration constant. This expresses a pulse with "area"

$$\int_{-\infty}^{\infty} \epsilon_L^2\, d\zeta = \frac{1}{\sqrt{C_3}}\left(\pi + 2\tan^{-1}\frac{C_2}{\sqrt{4C_1C_3}}\right). \tag{62b}$$

(b) $v_0 = 0$,

$$\epsilon_L^2 = \frac{C_2/C_3}{1 + (C_2^2/4C_3)(\zeta - \zeta_0)^2}. \tag{63a}$$

This is a Lorentzian pulse with area

$$\int_{-\infty}^{\infty} \epsilon_L^2\, d\zeta = \frac{2\pi}{\sqrt{C_3}}. \tag{63b}$$

The second type of solution (63) with $u_0 = 0$ and $w_0 = -1$ covers previous results. When $\omega_L = \omega_S$ and $Q_L = Q_S$, $\alpha_L^- = \alpha_S^-$, it agrees with the equal-frequency exact-resonance case of Belenov and Poluektov.[2] Tan-no *et al.*[3] obtained a Lorentzian pulse for different-frequency two-photon absorption neglecting the atomic frequency shift. Our result shows that the steady-state Lorentzian pulse is still valid with the frequency shift. The effect of the frequency shift is seen to be to loosen the pulse and decrease the pulse area.

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