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Theoretical study of photon echoes associated with the coherent nonlinear optical effect in a resonant three-level system

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Abstract. An analytical study of the photon echoes associated with the three-level system interacting resonantly with optical pulses possessing two different frequencies is performed. It is predicted that photon echoes arise at anomalous times corresponding to the correlation between the inhomogeneous broadenings for the different spectral lines, as well as at the normal time expected in a simple two-level system. This new effect is caused by the coupling of the two resonant transitions through the common energy level. Furthermore, if the particle does not have inversion symmetry, the echoes with the sum or difference frequency of the exciting optical pulses can be observed in such a system. The quantitative analysis is also given for intensities, direction of propagation and polarizations of the anomalous echoes which depend upon the exciting pulses with different frequency.

1. Introduction

The coherent interaction in a resonant nature between a many-particle system and optical waves has attracted new interest recently with the advent of coherent ultra-short light pulse techniques. In the case of the coherent and intense optical pulse shorter than the homogeneous relaxation time interacting with the many-particle quantum system, the change of the wavefunction caused by the interaction is so large that ordinary perturbation theory is no longer valid, and the dynamical interaction described by the exact treatment must be considered. At the same time, the coherence of the system plays an essential role in such a problem, since the phase memory of the wavefunction for each particle is maintained during the interaction.

This feature is primarily revealed by the photon echo (Abella *et al* 1966, Gordon *et al* 1969), the self-induced transparency (McCall and Hahn 1969, Lamb 1971) and the optical nutation (Tang and Silverman 1966) known as the new optical phenomena, and in particular, the photon echo is noteworthy as the typical example for the super-radiance (Bonifacio *et al* 1971a, b) first analysed by Dicke (1954).

However, studies of these phenomena have been mostly confined to those associated with the two-level system, that is, only the single frequency takes part in the problem. With respect to the problem where the optical waves with different frequencies are concerned, some theoretical approaches to the photon echo were carried out by Hartmann (1968), Nagibarov and Solvarov (1970), Tanno *et al* (1970) and Courtens

(1972). However, besides the Raman echo, these analyses did not take proper consideration of the correlation between the inhomogeneous broadenings for the different spectral lines. Even in the case of a three-level system, all possible transitions couple with each other because the raising or lowering operators for different transitions do not commute, and then photon echoes are formed at anomalous times, besides the normal time as expected from the usual analysis in the two-level system, depending upon correlation between the inhomogeneities of the different spectral lines. Furthermore, for the particle lacking inversion symmetry, we can expect echoes with the sum or difference frequencies of the exciting pulses caused by the nonlinearity which the three-level system possesses originally.

In this paper, we present the first part of the theoretical studies on the doubly resonant photon echoes in a three-level system using the density operator, in order to derive quantitatively their intensities, polarization characteristics and the propagation directions. Analytical descriptions are given in connection with not only the ordinary echoes corresponding to twice the time interval between successively applied excitation pulses, but also the anomalous echoes inherent to the coherent three-level system.

2. Equation of motion for the density operator in a three-level system

We can consider the system composed of many particles with three energy levels, which interact with coherent electromagnetic waves. The first problem to be considered is a system whose dimensions are small compared with the electromagnetic wavelengths. When dealing with this system, we must make use of the density operator for the whole system, since the photon echoes are the phenomena inherent to the many-particle system. However, if the distances between the particles are so large that the interaction between particles is negligible, the Liouville equation for the whole system can be separated into the equations for the individual particles. Then the density operator for the whole system can be obtained as the direct product of the individual particle density operators. Therefore, the starting point of our analysis is to solve the one-particle problem.

In this paper, we consider the case where the system is irradiated simultaneously by two coherent lights whose frequencies ω_{ba} and ω_{cb} are resonant at the centre frequencies of the inhomogeneously broadened spectral lines (figure 1). Since the dimensions of the system are small compared with the wavelength, the spatial dependence of the radiation fields can be omitted, and therefore the electric fields of the linearly polarized incident light can be described as

$$\begin{aligned}\boldsymbol{E}_{\mu\nu}(t) &= 2E_{\mu\nu 0}(t)(\boldsymbol{i}\cos\delta_{\mu\nu}+\boldsymbol{j}\sin\delta_{\mu\nu})\cos\omega_{\mu\nu}t \\ &= E_{\mu\nu 0}(t)\{\boldsymbol{i}\cos(\omega_{\mu\nu}t+\delta_{\mu\nu})+\boldsymbol{j}\sin(\omega_{\mu\nu}t+\delta_{\mu\nu})\}+E_{\mu\nu 0}(t)\{\boldsymbol{i}\cos(-\omega_{\mu\nu}t+\delta_{\mu\nu}) \\ &\qquad +\boldsymbol{j}\sin(-\omega_{\mu\nu}t+\delta_{\mu\nu})\},\end{aligned} \tag{1}$$

where subscript $\mu\nu$ denotes ba and cb, $\omega_{\mu\nu}$ is the frequency of the incident lights, $\boldsymbol{i}$ and $\boldsymbol{j}$ are the unit vectors along the spatial coordinate common to all particles, $\delta_{\mu\nu}$ is the angle between the directions of $\boldsymbol{i}$ and the incident electric fields, and $E_{\mu\nu 0}(t)$ is its envelope function. Equation (1) shows that the linearly polarized light can be expressed by the superposition of the right- and left-hand circularly polarized lights, and these are convenient for deriving the echo formula.

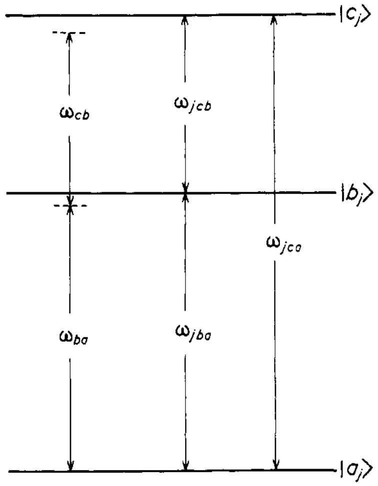

Figure 1. Schematic diagram of three-level system for the jth particle. Incident optical pulses with frequencies ω_{ba} and ω_{cb} are off-resonant because the energy levels are broadened inhomogeneously around the centre frequencies Ω_{ba0} and Ω_{cb0}, respectively.

The hamiltonian for a three-level system interacting with common radiation fields expressed by equation (1) can be written as

$$H = H_0 + H'_{ba} + H'_{cb} \tag{2}$$

where

$$H_0 = -\omega_{ba}|a\rangle\langle a| + \omega_{cb}|c\rangle\langle c|, \tag{3}$$

$$H'_{\mu\nu} = -\boldsymbol{p}_{\mu\nu}\cdot\boldsymbol{E}_{\mu\nu}(t) = -\sqrt{2}P_{\mu\nu}(R_{\mu\nu1}\boldsymbol{i} + R_{\mu\nu2}\boldsymbol{j})\cdot\boldsymbol{E}_{\mu\nu}(t). \tag{4}$$

Here $P_{\mu\nu}$ is the magnitude of the matrix element of the electric dipole moment operator with respect to the transitions a–b and b–c, and $R_{\mu\nu1}$ and $R_{\mu\nu2}$ are the Dicke operators for each transition, where the subscript to distinguish individual particles is omitted in order not to overburden the notation. These operators take the following forms in terms of the energy eigenstates:

$$\begin{aligned} R_{\mu\nu1} &= \frac{u_{\mu\nu}}{2}|\mu\rangle\langle\nu| + \frac{u^*_{\mu\nu}}{2}|\nu\rangle\langle\mu|, \\ R_{\mu\nu2} &= \frac{v_{\mu\nu}}{2}|\mu\rangle\langle\nu| + \frac{v^*_{\mu\nu}}{2}|\nu\rangle\langle\mu|. \end{aligned} \tag{5}$$

Parameters u and v are taken to be $u = 1$, $v = -\mathrm{i}$ for a σ_+ transition, and $u = 1$ and $v = \mathrm{i}$ for a σ_- transition. It is convenient to introduce the raising and lowering operators for each transition defined by

$$R_{\mu\nu\pm} = R_{\mu\nu1} \pm \mathrm{i}R_{\mu\nu2}. \tag{6}$$

Then the interaction part of the hamiltonian given by equation (4) can be rewritten as

$$H'_{\mu\nu} = -\frac{1}{\sqrt{2}}P_{\mu\nu}E_{\mu\nu0}(t)\{R_{\mu\nu+}\exp(-\mathrm{i}\delta_{\mu\nu}) + R_{\mu\nu-}\exp(\mathrm{i}\delta_{\mu\nu})\}\{\exp(\mathrm{i}\omega_{\mu\nu}t) + \exp(-\mathrm{i}\omega_{\mu\nu}t)\}. \tag{7}$$

A particle occupying three energy levels is generally described by a density operator $\sigma(t)$ which satisfies the equation of motion

$$i\hbar\frac{d\sigma(t)}{dt} = [H, \sigma(t)]. \tag{8}$$

It is convenient here to transform the density operator $\sigma(t)$ in the Schrödinger picture into the intermediate picture, but not the interaction picture, as follows:

$$\rho(t) = \exp(iSt)\sigma(t)\exp(-iSt), \tag{9}$$

where

$$S = \omega_{cb}|c\rangle\langle c| - \omega_{ba}|a\rangle\langle a|. \tag{10}$$

This unitary transformation leads to the equation of motion for $\rho(t)$

$$i\hbar\frac{d\rho(t)}{dt} = [\hbar\Delta + H_s', \rho(t)]. \tag{11}$$

where

$$\hbar\Delta = H_0 - \hbar S, \tag{12}$$

$$H_s' = \exp(iSt)(H_{ba}' + H_{cb}')\exp(-iSt). \tag{13}$$

Also Δ is diagonal in the energy representation with the eigenvalues $\Delta\omega_{\mu\nu} = \Omega_{\mu\nu} - \omega_{\mu\nu}$, and these values represent the measure of off-resonance between the incident radiation and the individual particle with transition frequency $\Omega_{\mu\nu}$ due to the inhomogeneous broadening of the spectral lines.

We should notice that equation (11) corresponds to the equation of motion in the rotating frame in the classical model, being used in the interpretation of spin as well as photon echoes in a two-level system. However, in the case of the three-level system, S represents the operator in the three-dimensional Hilbert space and contains the two different angular frequencies ω_{ba} and ω_{cb}.

In order to calculate equation (13), we have to derive the expression of $R_{\mu\nu\pm}$ in the new representation, as follows:

$$\begin{aligned} e^{iSt}R_{\mu\nu\pm}\,e^{-iSt} &= e^{iSt}\{\tfrac{1}{2}(u_{\mu\nu} + iv_{\mu\nu})|\mu\rangle\langle\nu| + \tfrac{1}{2}(u_{\mu\nu}^* \pm iv_{\mu\nu}^*)|\nu\rangle\langle\mu|\}\,e^{-iSt} \\ &= \tfrac{1}{2}(u_{\mu\nu} \pm iv_{\mu\nu})\exp(i\omega_{\mu\nu}t)|\mu\rangle\langle\nu| + \tfrac{1}{2}(u_{\mu\nu}^* \pm iv_{\mu\nu}^*)\exp(-i\omega_{\mu\nu}t)|\nu\rangle\langle\mu|. \end{aligned} \tag{14}$$

From the above equations, we obtain

$$\begin{aligned} e^{iSt}H_{\mu\nu}'\,e^{-iSt} = &-\frac{1}{2\sqrt{2}}P_{\mu\nu}E_{\mu\nu 0}(t)\{(u_{\mu\nu} + iv_{\mu\nu})\exp(-i\delta_{\mu\nu}) + (u_{\mu\nu} - iv_{\mu\nu})\exp(i\delta_{\mu\nu})\} \\ &\times \{1 + \exp(2i\omega_{\mu\nu}t)\}|\mu\rangle\langle\nu| + \text{hermitian adjoint}. \end{aligned} \tag{15}$$

We now make the rotating wave approximation, that is, we neglect the second term in the second braces in the above equation, which changes twice as rapidly as the radiation frequencies, since the component of $\rho(t)$ with slow time dependence compared to $\omega_{\mu\nu}^{-1}$ is of interest in our problem. This approximation means that, for the σ_+ (σ_-) transition, only the first (second) term in equation (1) interacts with them.

First we consider the case where the transition a–b is σ_+, and transition b–c is σ_-. Hence, equations (16) and (17) become simply

$$e^{iSt}H'_{ba}\,e^{-iSt} = -\frac{1}{\sqrt{2}}P_{ba}E_{ba0}(t)\{\exp(-i\delta_{ba})|b\rangle\langle a| + \exp(i\delta_{ba})|a\rangle\langle b|\}, \quad (16)$$

$$e^{iSt}H'_{cb}\,e^{-iSt} = -\frac{1}{\sqrt{2}}P_{cb}E_{cb0}(t)\{\exp(i\delta_{cb})|c\rangle\langle b| + \exp(-i\delta_{cb})|b\rangle\langle c|\}. \quad (17)$$

Introducing the operator T defined by

$$T = -\delta_{ba}|a\rangle\langle a| - \delta_{cb}|c\rangle\langle c|, \quad (18)$$

equations (16) and (17) are combined, then the interaction hamiltonian in the intermediate picture takes the form

$$H'_s = e^{-iT}\{-\sqrt{2}P_{ba}E_{ba0}(t)R_{ba1} - \sqrt{2}P_{cb}E_{cb0}(t)R_{cb1}\}\,e^{iT}. \quad (19)$$

3. Solution of the equation of motion for the density operator in a three-level system

In the previous section we have derived the equation of motion for the density operator which describes the dynamical behaviour of the particle occupying the three energy levels and interacting simultaneously with the incident waves whose frequencies are resonant to the line centre for each transition. We will try to solve the equation for the case where the system is excited by intense coherent optical pulses. For sufficiently intense exciting pulses it can be assumed that the off-resonance effect due to the inhomogeneous broadening of the spectral lines may be much smaller than the time evolution of the system caused by the interaction described by H'_s. Consequently, we ignore Δ during the duration of optical pulses to obtain

$$i\hbar\frac{d\rho(t)}{dt} = [H'_s, \rho(t)]. \quad (20)$$

In the period of absence of optical pulses, we have

$$i\hbar\frac{d\rho(t)}{dt} = [\hbar\Delta, \rho(t)]. \quad (21)$$

If we put $E_{cb0}(t) = kE_{ba0}(t)$, where k is an arbitrary time-independent constant, equation (20) can be solved exactly. This condition can be nearly achieved experimentally as follows: The N_2 laser beam, for example, is divided into two beams by a beam splitter in order to pump the two dye lasers which are tuned independently. The case where this condition is violated will be considered later.

For the optical pulses lasting from t_0 to $t_0+\tau$, the exact solution is expressed by

$$\begin{aligned}\rho(t_0+\tau) &= \exp\left(-\frac{i}{\hbar}\int_{t_0}^{t_0+\tau} H'_s\,dt\right)\rho(t_0)\exp\left(\frac{i}{\hbar}\int_{t_0}^{t_0+\tau} H'_s\,dt\right)\\ &= \exp[e^{-iT}\{i(\theta R_{ba1}+\phi R_{cb1})\}\,e^{iT}]\rho(t_0)\exp[e^{-iT}\{-i(\theta R_{ba1}+\phi R_{cb1})\}\,e^{iT}]\\ &= e^{-iT}\exp\{i(\theta R_{ba1}+\phi R_{cb1})\}\,e^{iT}\rho(t_0)\,e^{-iT}\exp\{-i(\theta R_{ba1}+\phi R_{cb1})\}\,e^{iT}, \end{aligned} \quad (22)$$

where

$$\theta = \frac{\sqrt{2}P_{ba}}{\hbar}\int_{t_0}^{t_0+\tau} E_{ba0}(t)\,\mathrm{d}t,$$
$$\phi = \frac{\sqrt{2}P_{cb}}{\hbar}\int_{t_0}^{t_0+\tau} E_{cb0}(t)\,\mathrm{d}t. \tag{23}$$

One should note that, if $E_{ba0}(t)/E_{cb0}(t)$ depends on time, solution (22) is no longer valid because of the fact that the unequal-time commutator bracket of H_s' does not vanish.

It is convenient to introduce the unitary operator

$$V = \frac{\theta_0}{\sqrt{2}}|a\rangle\langle a| - \phi_0|a\rangle\langle b| + \frac{\theta_0}{\sqrt{2}}|a\rangle\langle c| - \frac{1}{\sqrt{2}}|b\rangle\langle a| + \frac{1}{\sqrt{2}}|b\rangle\langle c| + \frac{\phi_0}{\sqrt{2}}|c\rangle\langle a| + \theta_0|c\rangle\langle b| + \frac{\phi_0}{\sqrt{2}}|c\rangle\langle c|$$

to rewrite equation (22)

$$\rho(t_0+\tau) = \mathrm{e}^{-\mathrm{i}T}V\exp\{\tfrac{1}{2}\mathrm{i}\psi(-|a\rangle\langle a|+|c\rangle\langle c|)\}V^{\dagger}\,\mathrm{e}^{\mathrm{i}T}\rho(t_0) \times \mathrm{e}^{-\mathrm{i}T}V\exp\{-\tfrac{1}{2}\mathrm{i}\psi(-|a\rangle\langle a|+|c\rangle\langle c|)\}V^{\dagger}\,\mathrm{e}^{\mathrm{i}T} \tag{24}$$

where $\psi = (\theta^2+\phi^2)^{1/2}$, $\theta_0 = \theta/(\theta^2+\phi^2)^{1/2}$ and $\phi_0 = \phi/(\theta^2+\phi^2)^{1/2}$. On the other hand, equation (21) can be solved simply as

$$\rho(t) = \exp\{-\mathrm{i}\Delta(t-t')\}\rho(t')\exp\{\mathrm{i}\Delta(t-t')\}. \tag{25}$$

Since unitary operator $\exp\{-\mathrm{i}\Delta(t-t')\}$ is diagonal, before or after the applied pulses the diagonal elements of $\rho(t)$ do not change and their off-diagonal elements change their phase with time at a different rate for each particle because the individual particles have the different eigenvalues of the off-resonance operator Δ.

4. Matrix elements of the density operator for discussion of the photon echoes associated with a three-level system

We consider the system irradiated by a sequence of two simultaneous optical pulses with different frequencies ω_{ba} and ω_{cb} separated by the same time interval τ_s, as shown in figure 2. Using the solutions (24) and (25), we derive the expression of the density operator at the time $t > \tau_s$, as follows:

$$\rho(t) = U\rho(0)U^{\dagger}. \tag{26}$$

Here the unitary operator U takes the form

$$U = \exp\left(-\frac{\mathrm{i}\Delta(t-\tau_s)}{\hbar}\right)\mathrm{e}^{-\mathrm{i}T'}V'\exp\{\tfrac{1}{2}\mathrm{i}\psi'(-|a\rangle\langle a|+|c\rangle\langle c|)\}V'^{\dagger}\,\mathrm{e}^{\mathrm{i}T'} \times\exp\left(-\frac{\mathrm{i}\Delta\tau_s}{\hbar}\right)\mathrm{e}^{-\mathrm{i}T}V\exp\{\tfrac{1}{2}\mathrm{i}\psi(-|a\rangle\langle a|+|c\rangle\langle c|)\}V^{\dagger}\,\mathrm{e}^{\mathrm{i}T}, \tag{27}$$

where the prime on operators denotes those for second pulses. Here we suppose that all particles are initially in the ground state $|a\rangle$, that is, $\rho(0) = |a\rangle\langle a|$.

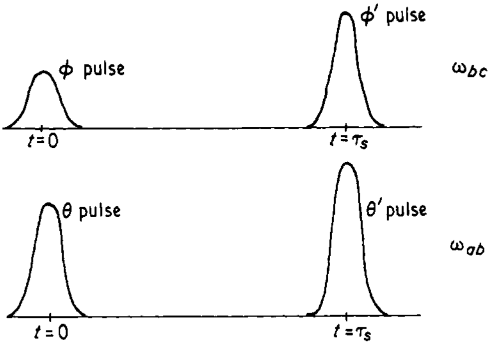

Figure 2. Schematic representation of two pulse sequencies with the frequencies ω_{ba} and ω_{cb} resonant to the transitions a–b and b–c in the three-level system, respectively.

We calculate the matrix elements of $\rho(t)$ in the energy representation. The diagonal elements of U take the form

$$\begin{aligned}\langle\mu|\rho|\mu\rangle &= \langle\mu|U|a\rangle\langle a|U^{\dagger}|\mu\rangle \\ &= |\langle\mu|U|a\rangle|^2 \qquad (\mu = a, b, c),\end{aligned} \tag{28}$$

and the off-diagonal elements are given by

$$\begin{aligned}\langle\mu|\rho|\nu\rangle &= \langle\mu|U|a\rangle\langle a|U^{\dagger}|\nu\rangle \\ &= \langle\mu|U|a\rangle\langle\nu|U|a\rangle^* \qquad (\mu, \nu = a, b, c).\end{aligned} \tag{29}$$

The matrix element of U can be calculated from equation (27), as follows:

$$\begin{aligned}\langle a|U|a\rangle = {} & (\theta_0^2\cos\tfrac{1}{2}\psi+\phi_0^2)(\theta_0'^2\cos\tfrac{1}{2}\psi'+\phi_0'^2)\exp(\mathrm{i}\Delta\omega_{ba}t)-\theta_0\theta_0'\sin\tfrac{1}{2}\psi\sin\tfrac{1}{2}\psi' \\ & \times\exp\{\mathrm{i}(\delta'_{ba}-\delta_{ba})\}\exp\{\mathrm{i}\Delta\omega_{ba}(t-\tau_s)\}+\theta_0\phi_0\theta_0'\phi_0'(1-\cos\tfrac{1}{2}\psi)(1-\cos\tfrac{1}{2}\psi') \\ & \times\exp\{\mathrm{i}(\delta'_{ba}-\delta_{ba}-\delta'_{cb}+\delta_{cb})\}\exp\{\mathrm{i}\Delta\omega_{ba}t-\mathrm{i}(\Delta\omega_{ba}+\Delta\omega_{cb})\tau_s\},\end{aligned} \tag{30a}$$

$$\begin{aligned}\langle b|U|a\rangle = {} & \mathrm{i}\theta_0'(\theta_0^2\cos\tfrac{1}{2}\psi+\phi_0^2)\sin\tfrac{1}{2}\psi'\exp(-\mathrm{i}\delta_{ba})\exp(\mathrm{i}\Delta\omega_{ba}\tau_s)+\mathrm{i}\theta_0\sin\tfrac{1}{2}\psi\cos\tfrac{1}{2}\psi' \\ & \times\exp(-\mathrm{i}\delta_{ba})+\mathrm{i}\theta_0\phi_0\phi_0'(1-\cos\tfrac{1}{2}\psi)\sin\tfrac{1}{2}\psi'\exp\{-\mathrm{i}(\delta_{ba}+\delta'_{cb}-\delta_{cb})\} \\ & \times\exp(-\mathrm{i}\Delta\omega_{cb}\tau_s)\end{aligned} \tag{30b}$$

and

$$\begin{aligned}\langle c|U|a\rangle = {} & \theta_0'\phi_0'(\theta_0^2\cos\tfrac{1}{2}\psi+\phi_0^2)(1-\cos\tfrac{1}{2}\psi')\exp\{\mathrm{i}(\delta'_{cb}-\delta'_{ba})\} \\ & \times\exp\{-\mathrm{i}\Delta\omega_{cb}t+\mathrm{i}(\Delta\omega_{ba}+\Delta\omega_{cb})\tau_s\}-\theta_0\phi_0'\sin\tfrac{1}{2}\psi\sin\tfrac{1}{2}\psi'\exp\{\mathrm{i}(\delta'_{cb}-\delta_{ba})\} \\ & \times\exp\{-\mathrm{i}\Delta\omega_{cb}(t-\tau_s)\}+\theta_0\phi_0(1-\cos\tfrac{1}{2}\psi)(\theta_0'^2+\phi_0'^2\cos\tfrac{1}{2}\psi') \\ & \times\exp\{\mathrm{i}(\delta_{cb}-\delta_{ba})\}\exp(-\mathrm{i}\Delta\omega_{cb}t),\end{aligned} \tag{30c}$$

where the relation $|a\rangle\langle a|+|b\rangle\langle b|+|c\rangle\langle c| = I$ (I is the unit operator in the three-dimensional Hilbert space) was used. Substitution of equations (30a)–(30c) into equation

(29) gives

$$\langle a|\rho|b\rangle = \mathrm{i}A_{2\tau_s}A'_{2\tau_s}\exp\{\mathrm{i}(2\delta'_{ba}-\delta_{ba})\}\exp\{\mathrm{i}\Delta\omega_{ba}(t-2\tau_s)\}-\mathrm{i}A_{(2+\beta)\tau_s}A'_{(2+\beta)\tau_s}$$
$$\times\exp\{\mathrm{i}(2\delta'_{ba}-\delta_{ba}-\delta'_{cb}+\delta_{cb})\}\exp\{\mathrm{i}\Delta\omega_{ba}t-(2\Delta\omega_{ba}+\Delta\omega_{cb})\tau_s\}$$
$$-\mathrm{i}A_{(1+\beta)\tau_s}A'_{(1+\beta)\tau_s}\exp\{\mathrm{i}(\delta'_{ba}-\delta'_{cb}+\delta_{cb})\}\exp\{\mathrm{i}\Delta\omega_{ba}t-\mathrm{i}(\Delta\omega_{ba}+\Delta\omega_{cb})\tau_s\}$$
$$+\mathrm{i}A_{(1-\beta)\tau_s}A'_{(1-\beta)\tau_s}\exp\{\mathrm{i}(\delta'_{ba}+\delta'_{cb}-\delta_{cb})\}\exp\{\mathrm{i}\Delta\omega_{ba}t-\mathrm{i}(\Delta\omega_{ba}-\Delta\omega_{cb})\tau_s\}$$
$$-\mathrm{i}A_{-\beta\tau_s}A'_{-\beta\tau_s}\exp\{\mathrm{i}(\delta_{ba}+\delta'_{cb}-\delta_{cb})\}\exp(\mathrm{i}\Delta\omega_{ba}t+\mathrm{i}\Delta\omega_{cb}\tau_s), \tag{31a}$$

$$\langle b|\rho|c\rangle = -\mathrm{i}B_{2\tau_s}B'_{2\tau_s}\exp\{-\mathrm{i}(2\delta'_{cb}-\delta_{cb})\}\exp\{\mathrm{i}\Delta\omega_{cb}(t-2\tau_s)\}+\mathrm{i}B_{(2+\beta^{-1})\tau_s}B'_{(2+\beta^{-1})\tau_s}$$
$$\times\exp\{-\mathrm{i}(2\delta'_{cb}-\delta_{cb}-\delta'_{ba}+\delta_{ba})\}\exp\{\mathrm{i}\Delta\omega_{cb}t-\mathrm{i}(\Delta\omega_{ba}+2\Delta\omega_{cb})\tau_s\}$$
$$+\mathrm{i}B_{(1+\beta^{-1})\tau_s}B'_{(1+\beta^{-1})\tau_s}\exp\{-\mathrm{i}(\delta'_{cb}-\delta'_{ba}+\delta_{ba})\}\exp\{\mathrm{i}\Delta\omega_{cb}t$$
$$-\mathrm{i}(\Delta\omega_{ba}+\Delta\omega_{cb})\tau_s\}-\mathrm{i}B_{(1-\beta^{-1})\tau_s}B'_{(1-\beta^{-1})\tau_s}\exp\{-\mathrm{i}(\delta'_{cb}+\delta'_{ba}-\delta_{ba})\}$$
$$\times\exp\{\mathrm{i}\Delta\omega_{cb}\mathrm{t}-\mathrm{i}(\Delta\omega_{ba}+\Delta\omega_{cb})\tau_s\}+\mathrm{i}B_{-\beta^{-1}\tau_s}B'_{-\beta^{-1}\tau_s}$$
$$\times\exp\{-\mathrm{i}(\delta_{cb}+\delta'_{ba}-\delta_{ba})\}\exp(\mathrm{i}\Delta\omega_{cb}t+\mathrm{i}\Delta\omega_{ba}\tau_s) \tag{31b}$$

where $\beta = \alpha_{cb}/\alpha_{ba}$,

$$A_{2\tau_s} = \theta_0\sin\tfrac{1}{2}\psi(\theta_0^2\cos\tfrac{1}{2}\psi+\phi_0^2), \tag{32a}$$
$$A'_{2\tau_s} = \theta_0'^2\sin^2\tfrac{1}{2}\psi', \tag{32b}$$
$$A_{(2+\beta)\tau_s} = \theta_0\phi_0(1-\cos\tfrac{1}{2}\psi)(\theta_0^2\cos\tfrac{1}{2}\psi+\phi_0^2), \tag{32c}$$
$$A'_{(2+\beta)\tau_s} = \theta_0'^2\phi_0'\sin\tfrac{1}{2}\psi'(1-\cos\tfrac{1}{2}\psi'), \tag{32d}$$
$$A_{(1+\beta)\tau_s} = \theta_0^2\phi_0\sin\tfrac{1}{2}\psi(1-\cos\tfrac{1}{2}\psi), \tag{32e}$$
$$A'_{(1+\beta)\tau_s} = \theta_0'\phi_0'\cos\tfrac{1}{2}\psi'(1-\cos\tfrac{1}{2}\psi'), \tag{32f}$$
$$A_{(1-\beta)\tau_s} = A_{(1+\beta)\tau_s}, \tag{32g}$$
$$A'_{(1-\beta)\tau_s} = \theta_0'\phi_0'\sin^2\tfrac{1}{2}\psi', \tag{32h}$$
$$A_{-\beta t_s} = A_{(2+\beta)\tau_s}, \tag{32i}$$
$$A'_{-\beta\tau_s} = \theta_0'\sin\tfrac{1}{2}\psi'(\theta_0'^2\cos\tfrac{1}{2}\psi'+\phi_0'^2), \tag{32j}$$
$$B_{2\tau_s} = A_{(1+\beta)\tau_s}, \tag{32k}$$
$$B'_{2\tau_s} = \phi_0'^2\sin^2\tfrac{1}{2}\psi', \tag{32l}$$
$$B_{(2+\beta^{-1})\tau_s} = A_{(2+\beta)\tau_s}, \tag{32m}$$
$$B'_{(2+\beta^{-1})\tau_s} = \theta_0'\phi_0'^2\sin\tfrac{1}{2}\psi'(1-\cos\tfrac{1}{2}\psi'), \tag{32n}$$
$$B_{(1+\beta^{-1})\tau_s} = A_{2\tau_s}, \tag{32o}$$
$$B'_{(1+\beta^{-1})\tau_s} = A'_{(1+\beta)\tau_s}, \tag{32p}$$
$$B_{(1-\beta^{-1})\tau_s} = A_{2\tau_s}, \tag{32q}$$
$$B'_{(1-\beta^{-1})\tau_s} = A'_{(1-\beta)\tau_s}, \tag{32r}$$
$$B_{-\beta^{-1}\tau_s} = A_{(2+\beta)\tau_s}, \tag{32s}$$
$$B'_{-\beta^{-1}\tau_s} = \theta_0'\sin\tfrac{1}{2}\psi'(\theta_0'^2+\phi_0'^2\cos\tfrac{1}{2}\psi'), \tag{32t}$$

and we omitted the terms which do not contribute to the echo formation. As will be apparent in the next section, the off-diagonal elements of the density operator obtained above correspond directly to the expectation values of the electric-dipole moment operator, so that they determine the characteristics of the photon echoes. On the other hand, the diagonal elements are not essential in our problem. The physical meaning of β in equations (31*a*)–(32*t*) will also become apparent in the next section.

One should note that equations (31*a*), (31*b*) suggest that the photon echo is produced not only at $t = 2\tau_s$, that is, normal time, but also at several anomalous times which depend on the ratio $\Delta\omega_{ba}/\Delta\omega_{cb}$. In the case where exciting pulses are exactly resonant at the centres of the inhomogeneously broadened lines, $\Delta\omega_{\mu\nu}$ can be written approximately

$$\Delta\omega_{\mu\nu} = \alpha_{\mu\nu}x \tag{32}$$

where x is the physical parameter determined by taking account of the cause of the inhomogeneous broadening of lines. For example, in the case of gases, x describes the component of the velocity of a particle along the line of sight, and in the case of solids, the deviation of the crystalline field at the position of the particle from its mean value. Moreover, in gases, the coefficients α_{ba} and α_{cb} in equation (32) can be written simply as $\alpha_{ba} = \Omega_{ba0}/c$ and $\alpha_{cb} = \Omega_{cb0}/c$, where Ω_{cb0} is the centre frequency and c is the light velocity. However, in solids, it is to be noted that such simple relations do not hold, and α_{ba} and α_{cb} can even take negative values in some cases.

5. Intensity and polarization characteristics of photon echoes

In this section we consider the echo intensity and polarization for the case where each energy level has twofold degeneracy and the exciting pulses are perpendicular to the quantization axis. In this case, the interaction process is described by the superposition of the two independent sets of transitions as shown in figure 3. This situation can be achieved by using, for example, the transitions $n^2S_{1/2} - n'^2P_{1/2} - n''^2S_{1/2}$ (or $-n''^2D_{1/2}$) of an alkali vapour or those between the Kramers doublets of R and B lines of the ruby crystal.

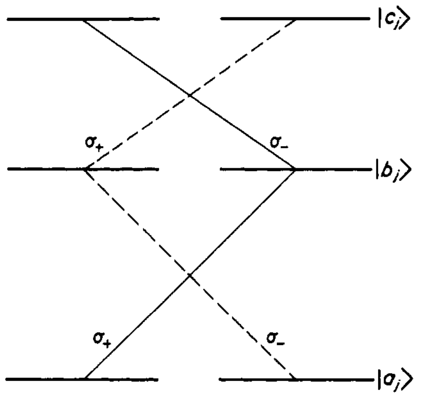

Figure 3. Schematic representation of the three-level system where each level has twofold degeneracy. Full and broken lines show the two independent sets of transitions caused by the linearly polarized radiation perpendicular to the quantization axis.

In order to find the echo intensity and polarization, we have to obtain the expectation values of the electric-dipole moment operators for each transition and to integrate them over the inhomogeneous broadenings of the spectral lines. When performing this, one should note that the two off-resonance parameters $\Delta\omega_{ba}$ and $\Delta\omega_{cb}$ take part in the problem unlike the conventional two-level system. However, $\Delta\omega_{ba}$ and $\Delta\omega_{cb}$ are not independent from each other, and are related to the physical parameter x causing the inhomogeneous broadening as shown in equation (32). Therefore, the total electric-dipole moment exhibited by an ensemble of particles is expressed by

$$\begin{aligned}\langle \boldsymbol{p}_{\mu\nu}(t)\rangle &= N\int_{-\infty}^{\infty} g(x)\,\mathrm{Tr}(\boldsymbol{p}_{\mu\nu}\sigma(t))\,\mathrm{d}x \\ &= P_{\mu\nu}N\int_{-\infty}^{\infty} g(x)\left(\frac{1}{\sqrt{2}}(\boldsymbol{i}-\mathrm{i}\boldsymbol{j})\langle\nu|\rho|\mu\rangle\exp(-\mathrm{i}\omega_{\mu\nu}t)+\mathrm{cc}\right)\mathrm{d}x \qquad (33)\end{aligned}$$

where N is the number of particles and $g(x)$ is a distribution function for x. It is to be noted that the electric-dipole moment at the frequency ω_{ca} is polarized along the quantization axis, that is, perpendicular to the direction of the electric fields of the excitation pulses. Accordingly, our interest is only limited to evaluating equation (33) for the dipole moments with frequencies ω_{ba} and ω_{cb}.

We have already obtained the matrix elements of the density operator, $\langle a|\rho|b\rangle$ and $\langle b|\rho|c\rangle$, for the set of transitions shown by the full lines in figure 3. In the same fashion we can also obtain those for the broken lines in figure 3. Substituting the density operator for each set of transitions into equation (33) independently, and superposing these results, we obtain

$$\begin{aligned}\langle \boldsymbol{p}_{ba}\rangle = \sqrt{2}P_{ba}N[&-A_{2\tau_s}A'_{2\tau_s}G(\alpha_{ba}(t-2\tau_s))\{\boldsymbol{i}\cos(2\delta'_{ba}-\delta_{ba})+\boldsymbol{j}\sin(2\delta'_{ba}-\delta_{ba})\} \\ &+A_{(2+\beta)\tau_s}A'_{(2+\beta)\tau_s}G(\alpha_{ba}\{t-(2+\beta)\tau_s\})\{\boldsymbol{i}\cos(2\delta'_{ba}-\delta_{ba}-\delta'_{cb}+\delta_{cb}) \\ &+\boldsymbol{j}\sin(2\delta'_{ba}-\delta_{ba}-\delta'_{cb}+\delta_{cb})\}+A_{(1+\beta)\tau_s}A'_{(1+\beta)\tau_s}G(\alpha_{ba}\{t-(1+\beta)\tau_s\}) \\ &\times\{\boldsymbol{i}\cos(\delta'_{ba}+\delta_{cb}-\delta'_{cb})+\boldsymbol{j}\sin(\delta'_{ba}+\delta_{cb}-\delta'_{cb})\}+A_{(1-\beta)\tau_s}A'_{(1-\beta)\tau_s} \\ &\times G(\alpha_{ba}\{t-(1-\beta)\tau_s\})\{\boldsymbol{i}\cos(\delta'_{ba}+\delta'_{cb}-\delta_{cb})+\boldsymbol{j}\sin(\delta'_{ba}+\delta'_{cb}-\delta_{cb})\} \\ &+A_{-\beta\tau_s}A'_{-\beta\tau_s}G(\alpha_{ba}(t+\beta\tau_s))\{\boldsymbol{i}\cos(\delta_{ba}+\delta'_{cb}-\delta_{cb})+\boldsymbol{j}\sin(\delta_{ba}+\delta'_{cb}-\delta_{cb})\}] \\ &\times\sin\omega_{ba}t, \qquad (34a)\end{aligned}$$

and

$$\begin{aligned}\langle \boldsymbol{p}_{cb}\rangle = \sqrt{2}P_{cb}N[&-B_{2\tau_s}B'_{2\tau_s}G\{\alpha_{cb}(t-2\tau_s)\}\{\boldsymbol{i}\cos(2\delta'_{cb}-\delta_{cb})+\boldsymbol{j}\sin(2\delta'_{cb}-\delta_{cb})\} \\ &+B_{(2+\beta^{-1})\tau_s}B'_{(2+\beta^{-1})\tau_s}G(\alpha_{cb}\{t-(2+\beta^{-1})\tau_s\})\{\boldsymbol{i}\cos(2\delta'_{cb}-\delta_{cb}-\delta'_{ba}+\delta_{ba}) \\ &+\boldsymbol{j}\sin(2\delta'_{cb}-\delta_{cb}-\delta'_{ba}+\delta_{ba})\}+B_{(1+\beta^{-1})\tau_s}B'_{(1+\beta^{-1})\tau_s}G(\alpha_{cb}\{t-(1+\beta^{-1})\tau_s\}) \\ &\times\{\boldsymbol{i}\cos(\delta'_{cb}+\delta_{ba}-\delta'_{ba})+\boldsymbol{j}\sin(\delta'_{cb}+\delta_{ba}-\delta'_{ba})\}+B_{(1-\beta^{-1})\tau_s}B'_{(1-\beta^{-1})\tau_s} \\ &\times G(\alpha_{cb}\{t-(1-\beta^{-1})\tau_s\})\{\boldsymbol{i}\cos(\delta'_{cb}+\delta'_{ba}-\delta_{ba})+\boldsymbol{j}\sin(\delta'_{cb}+\delta'_{ba}-\delta_{ba})\} \\ &+B_{-\beta^{-1}\tau_s}B'_{-\beta^{-1}\tau_s}G(\alpha_{cb}(t+\beta^{-1}\tau_s))\{\boldsymbol{i}\cos(\delta_{cb}+\delta'_{ba}-\delta_{ba}) \\ &+\boldsymbol{j}\sin(\delta_{cb}+\delta'_{ba}-\delta_{ba})\}]\sin\omega_{cb}t, \qquad (34b)\end{aligned}$$

where $\beta = \alpha_{cb}/\alpha_{ba}$. As is evident from the above equations, the shapes of echoes are expressed by the Fourier transform of the distribution function of x, which is given by

$$G(\xi) = \int_{-\infty}^{\infty} g(x)\,\mathrm{e}^{-\mathrm{i}\xi x}\,\mathrm{d}x.$$

From equations (34*a*) and (34*b*), we obtained the remarkable results that the photon echoes are also produced at some anomalous times different from $t = 2\tau_s$. Equation (34*a*) shows that the two anomalous echoes with frequency ω_{ba} arise depending upon the following three cases: (i) in the case of $\beta > 0$, echoes appear at $(2+\beta)\tau_s$ and $(1+\beta)\tau_s$, besides at $2\tau_s$; (ii) in the case of $-1 < \beta < 0$, echoes appear at $2\tau_s$, $(2-|\beta|)\tau_s$ and $(1+|\beta|)\tau_s$; and (iii) in the case of $\beta < -1$, echoes arise at $2\tau_s$, $(1+|\beta|)\tau_s$ and $|\beta|\tau_s$. Equation (34*b*) shows that the two anomalous echoes with frequency ω_{cb} also appear: (i) in the case of $\beta > 0$, echoes appear at $2\tau_s$, $(2+\beta^{-1})\tau_s$ and $(1+\beta^{-1})\tau_s$; (ii) in the case of $-1 < \beta < 0$, echoes appear at $2\tau_s$, $(2-|\beta|^{-1})\tau_s$ and $(1+|\beta|^{-1})\tau_s$; (iii) in the case of $\beta < -1$, echoes appear at $2\tau_s$, $(1+|\beta|^{-1})\tau_s$ and $|\beta|^{-1}\tau_s$.

As mentioned previously, in the case where the inhomogeneous broadening is caused by the Doppler effect as in gases, only the first case is possible and the time when the echo arises is essentially determined by the ratio of energy separations since the relation $\beta = \Omega_{bc0}/\Omega_{ba0}$ holds. However, in solids, where the spatial fluctuation of the static crystalline field gives rise to the inhomogeneous broadening of energy levels, the variation of the energy eigenvalues with the magnitude of the crystalline field is complicated depending upon not only the electron configuration but the configuration interaction. Consequently, the echoes are produced at times different from the case of gases even if the ratio of the energy separations $\Omega_{cb0}/\Omega_{ba0}$ is the same. It is pointed out here for the first time, as far as we know, that this interesting feature in photon echoes is inherent to the three-level system, and can be explained as the effect caused by coherent coupling between the two transitions *a–b* and *b–c*. The detailed discussion of this feature will be made later.

We now consider the case that the limitation $E_{cb0}(t) = kE_{ba0}(t)$ does not exist. In such a case we can not obtain the analytic expression, and the results on the intensity and polarization characteristics of echoes can be obtained only by numerical integration. One should, however, note that our results on times when echoes arise are still valid, since they are determined only by β.

Furthermore, if the particle does not possess the inversion symmetry, echoes with the sum frequency $\omega_{ca} = \omega_{ba} + \omega_{cb}$ can be produced by considering the case where all three transitions are π transitions. Then the result is given by

$$\begin{aligned}\langle \boldsymbol{p}_{ca}\rangle = \sqrt{2}P_{ca}N[&-C_{2\tau_s}C'_{2\tau_s}G(\alpha_{ca}(t-2\tau_s)) + C_{\{1+(1+\beta)^{-1}\}\tau_s}C'_{\{1+(1+\beta)^{-1}\}\tau_s}\\ &\times G(\alpha_{ca}[t-\{1+(1+\beta)^{-1}\}\tau_s]) + C_{\{1+(1+\beta^{-1})^{-1}\}\tau_s}C'_{\{1+(1+\beta^{-1})^{-1}\}\tau_s}\\ &\times G(\alpha_{ca}[t-\{1+(1+\beta^{-1})^{-1}\}\tau_s]) + C_{(1+\beta)^{-1}\tau_s}C'_{(1+\beta)^{-1}\tau_s}\\ &\times G[\alpha_{ca}\{t-(1+\beta)^{-1}\tau_s\}]G(\alpha_{ca}\{t-(1+\beta)^{-1}\tau_s\}) + C_{(1+\beta^{-1})^{-1}\tau_s}C'_{(1+\beta^{-1})^{-1}\tau_s}\\ &\times G(\alpha_{ca}\{t-(1+\beta^{-1})^{-1}\}\tau_s)]\boldsymbol{k}\sin\omega_{ca}t, \end{aligned} \tag{35}$$

where

$$C_{2\tau_s} = A_{(2+\beta)\tau_s}, \tag{36a}$$

$$C'_{2\tau_s} = \theta_0'^2\phi_0'^2(1-\cos\tfrac{1}{2}\psi')^2, \tag{36b}$$

$$C_{\{1+(1+\beta)^{-1}\}\tau_s} = A_{2\tau_s}, \tag{36c}$$

$$C'_{\{1+(1+\beta)^{-1}\}\tau_s} = A'_{(2+\beta)\tau_s}, \tag{36d}$$

$$C_{\{1+(1+\beta^{-1})^{-1}\}\tau_s} = A_{(1+\beta)\tau_s}, \tag{36e}$$

$$C'_{\{1+(1+\beta^{-1})^{-1}\}\tau_s} = B'_{(2+\beta^{-1})\tau_s}, \tag{36f}$$

$$C_{(1+\beta)^{-1}\tau_s} = A_{(1+\beta)\tau_s}, \tag{36g}$$

$$C'_{(1+\beta)^{-1}\tau_s} = B'_{-\beta^{-1}\tau_s}, \tag{36h}$$

$$C_{(1+\beta^{-1})^{-1}\tau_s} = A_{2\tau_s}, \tag{36i}$$

$$C'_{(1+\beta^{-1})^{-1}t_s} = A'_{-\beta\tau_s}. \tag{36j}$$

To summarize the results on the intensity characteristics, table 1 shows the times of echo formation and the maximum intensities of the echoes at the frequencies ω_{ba}, ω_{cb} and ω_{ca}, depending upon the three cases as mentioned before. Figures 4(*a*)–(*d*) show some examples of the squared value of the coefficients A, B and C (A', B', and C') which indicate how the intensity of the echoes depends on the exciting pulse areas θ and ϕ (θ' and ϕ') for the first (second) pulses. We notice here that some of these coefficients have the same functional dependence or become identical by exchanging θ and ϕ, or θ' and ϕ', and thus only eight coefficients are found to be independent. Various intensity characteristics of the echo radiation involved in the three-level system can be found based on our analysis. For instance, figure 4(*a*) shows that the periodicity of the echo intensity collapses for the case of $\phi \neq 0$. Especially, the combination of the $\theta = 3\pi$ pulse and the $\phi = 1{\cdot}7\pi$ pulse induces the echo at the frequency ω_{ba} which can not be expected for the 3π pulse only in the simple two-level system, and its intensity becomes almost the same as the maximum value of the normal echo at $t = 2\tau_s$.

Table 1. Maximum values of the photon echo intensity with the frequencies ω_{ba}, ω_{cb} and ω_{ca} associated with a resonant three-level system being classified into three cases according to the value of β. $\beta = \alpha_{cb}/\alpha_{ba}$ is the parameter indicating the correlation between the inhomogeneous broadenings of the different spectral lines, and determines the times when echo arises. $I_{\mu\nu 0} = 4\omega_{\mu\nu}^3 P_{\mu\nu}^2/3\hbar c^3$

Frequency of echo	Time when echo arises	Maximum value of echo intensity†		
		$\beta > 0$	$-1 < \beta < 0$	$\beta < -1$
ω_{ba}	$2\tau_s$	$\frac{1}{4}N^2 I_{ab0}$	$\frac{1}{4}N^2 I_{ab0}$	$\frac{1}{4}N^2 I_{ab0}$
	$(2+\beta)\tau_s$	$\frac{1}{16}N^2 I_{ab0}$	$\frac{1}{16}N^2 I_{ab0}$	—
	$(1+\beta)\tau_s$	$\frac{1}{4}N^2 I_{ab0}$	—	—
	$(1-\beta)\tau_s$	—	$\frac{1}{16}N^2 I_{ab0}$	$\frac{1}{16}N^2 I_{ab0}$
	$-\beta\tau_s$	—	—	$\frac{1}{4}N^2 I_{ab0}$
ω_{cb}	$2\tau_s$	$\frac{1}{4}N^2 I_{bc0}$	$\frac{1}{4}N^2 I_{bc0}$	$\frac{1}{4}N^2 I_{bc0}$
	$(2+\beta^{-1})\tau_s$	$\frac{1}{16}N^2 I_{bc0}$	$\frac{1}{16}N^2 I_{bc0}$	—
	$(1+\beta^{-1})\tau_s$	$\frac{1}{4}N^2 I_{bc0}$	—	—
	$(1-\beta^{-1})\tau_s$	—	$\frac{1}{16}N^2 I_{bc0}$	$\frac{1}{16}N^2 I_{bc0}$
	$-\beta^{-1}\tau_s$	—	—	$\frac{1}{4}N^2 I_{bc0}$
ω_{ca}	$2\tau_s$	$\frac{1}{4}N^2 I_{ac0}$	$\frac{1}{4}N^2 I_{ac0}$	$\frac{1}{4}N^2 I_{ac0}$
	$\{1+(1+\beta)^{-1}\}\tau_s$	$\frac{1}{16}N^2 I_{ac0}$	$\frac{1}{16}N^2 I_{ac0}$	—
	$\{1+(1+\beta^{-1})^{-1}\}\tau_s$	$\frac{1}{16}N^2 I_{ac0}$	—	$\frac{1}{16}N^2 I_{ac0}$
	$(1+\beta)^{-1}\tau_s$	—	$\frac{1}{4}N^2 I_{ac0}$	—
	$(1+\beta^{-1})^{-1}\tau_s$	—	—	$\frac{1}{4}N^2 I_{ac0}$

‡ Subscripts *ab* and *bc* should read *ba* and *cb* respectively in entries under this heading.

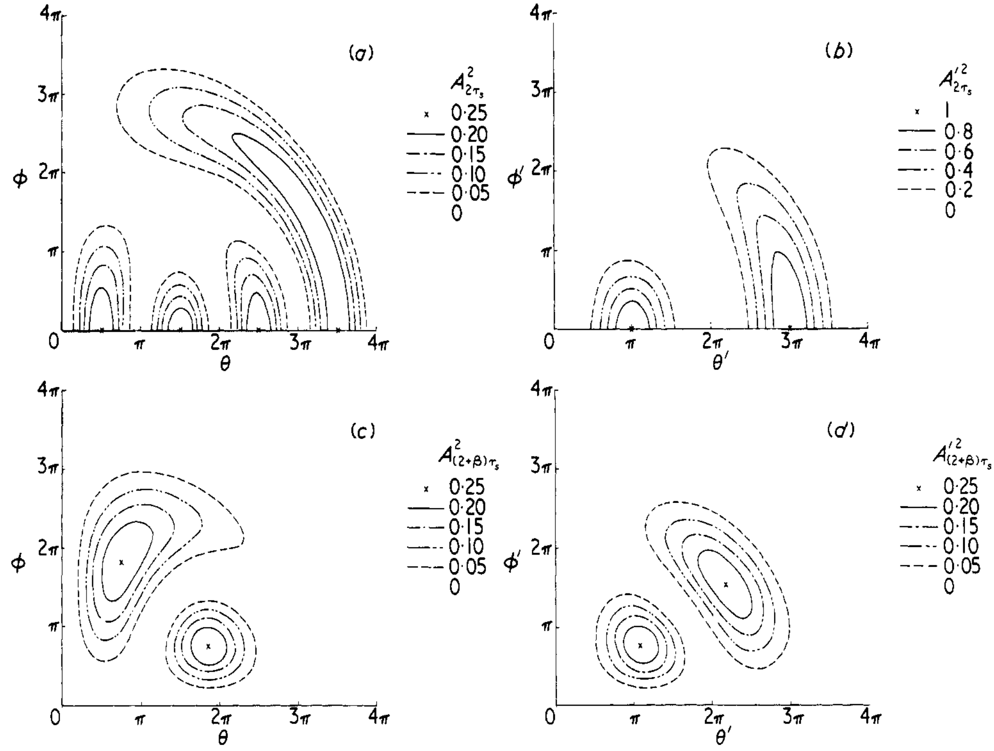

Figure 4. The distribution of the numerical values of $A^2_{2\tau_s}$, $A'^2_{2\tau_s}$, $A^2_{(2+\beta)\tau_s}$ and $A'^2_{(2+\beta)\tau_s}$ indicating the change of the intensity of photon echoes as a function of the areas of the incident optical pulses. (*a*) The value of $A^2_{2\tau_s}$ as a function of the areas for the first optical pulses, θ and ϕ. (*b*) The values of $A'^2_{2\tau_s}$ as a function of the areas for the second optical pulses, θ' and ϕ'. (*c*) The values of $A^2_{(2+\beta)\tau_s}$ as a function of the areas for the first optical pulses, θ and ϕ. (*d*) The value of $A'^2_{(2+\beta)\tau_s}$ as a function of the areas for the second optical pulses, θ' and ϕ'.

Equations (34*a*) and (34*b*) also show the polarization characteristics of the echo radiation. What is evident from these equations is that the polarization of the anomalous echoes is affected by the exciting pulses with frequency different from that of the echoes, while the polarization of the normal echo at $t = 2\tau_s$ agrees with the result given by Abella *et al* (1966) for the two-level system.

6. Propagation direction of photon echoes

Until the previous section, we analysed the echo formation in the three-level system whose dimension is small compared with the excitation wavelengths. This means that all particles feel the exciting optical fields at the same time.

In this section we consider the system with dimension larger than the wavelength, but smaller than the coherence length of the spontaneous radiation. In such a large volume case, we must take into account the fact that there exists a difference in arrival time of the pulses at the particles. That is, equation (22) should be rewritten as follows:

$$\rho(t_0+\tau) = \mathrm{e}^{-\mathrm{i}T}\exp\{\mathrm{i}(\theta R_{bak1}+\phi R_{cbk1})\}\,\mathrm{e}^{\mathrm{i}T}\rho(t_0)\,\mathrm{e}^{-\mathrm{i}T}\exp\{-\mathrm{i}(\theta R_{bak1}+\phi R_{cbk1})\}\,\mathrm{e}^{\mathrm{i}T}, \quad (37)$$

where

$$R_{bak1} = R_{ba1}\cos(\boldsymbol{k}_{ba}\cdot\boldsymbol{r}) - R_{ba2}\sin(\boldsymbol{k}_{ba}\cdot\boldsymbol{r}), \tag{38a}$$

$$R_{bak2} = R_{ba1}\sin(\boldsymbol{k}_{ba}\cdot\boldsymbol{r}) + R_{ba2}\cos(\boldsymbol{k}_{ba}\cdot\boldsymbol{r}), \tag{38b}$$

$$R_{cbk1} = R_{cb1}\cos(\boldsymbol{k}_{cb}\cdot\boldsymbol{r}) + R_{cb2}\sin(\boldsymbol{k}_{cb}\cdot\boldsymbol{r}), \tag{38c}$$

$$R_{cbk2} = -R_{cb1}\sin(\boldsymbol{k}_{cb}\cdot\boldsymbol{r}) + R_{cb2}\cos(\boldsymbol{k}_{cb}\cdot\boldsymbol{r}), \tag{38d}$$

and $\boldsymbol{k}_{ba}$ and $\boldsymbol{k}_{cb}$ are the wavevectors of the exciting pulses.

Assuming, for the sake of simplicity, that the polarization directions of the exciting pulses are all parallel, the total electric-dipole moment considering the phase difference among the particles can be written as

$$\langle \boldsymbol{p}_{\mu\nu k_{\mu\nu e}}(t)\rangle = \left\langle \int_{-\infty}^{\infty} g(x)\,\mathrm{Tr}(\boldsymbol{p}_{\mu\nu k_{\mu\nu e}}\sigma(t))\,\mathrm{d}x \right\rangle_{\mathrm{av}}, \tag{39}$$

where

$$\boldsymbol{p}_{\mu\nu k_{\mu\nu e}} = \sqrt{2}P_{\mu\nu}(R_{\mu\nu k_{\mu\nu e}1}\boldsymbol{i} + R_{\mu\nu k_{\mu\nu e}2}\boldsymbol{j}) \tag{40}$$

and $\langle \ldots \rangle_{\mathrm{av}}$ means the averaging over the spatial distribution of particles.

Hence we obtain

$$\begin{aligned}\langle \boldsymbol{p}_{bak_{bae}}\rangle = {} & \sqrt{2}P_{ba}N[-A_{2\tau_s}A'_{2\tau_s}G(\alpha_{ba}(t-2\tau_s))\langle\cos\{(\boldsymbol{k}_{bae}-2\boldsymbol{k}'_{ba}+\boldsymbol{k}_{ba})\cdot\boldsymbol{r}\}\rangle_{\mathrm{av}} + A_{(2+\beta)\tau_s} \\ & \times A'_{(2+\beta)\tau_s}G(\alpha_{ba}\{t-(2+\beta)\tau_s\})\langle\cos\{(\boldsymbol{k}_{bae}-2\boldsymbol{k}'_{ba}+\boldsymbol{k}_{ba}-\boldsymbol{k}'_{cb}+\boldsymbol{k}_{cb})\cdot\boldsymbol{r}\}\rangle_{\mathrm{av}} \\ & + A_{(1+\beta)\tau_s}A'_{(1+\beta)\tau_s}G(\alpha_{ba}\{t-(1+\beta)\tau_s\})\langle\cos\{(\boldsymbol{k}_{bae}-\boldsymbol{k}'_{ba}-\boldsymbol{k}'_{cb}+\boldsymbol{k}_{cb})\cdot\boldsymbol{r}\}\rangle_{\mathrm{av}} \\ & + A_{(1-\beta)\tau_s}A'_{(1-\beta)\tau_s}G(\alpha_{ba}\{t-(1-\beta)\tau_s\})\langle\cos\{(\boldsymbol{k}_{bae}-\boldsymbol{k}'_{ba}+\boldsymbol{k}'_{cb}-\boldsymbol{k}_{cb})\cdot\boldsymbol{r}\}\rangle_{\mathrm{av}} \\ & + A_{-\beta\tau_s}A'_{-\beta\tau_s}G(\alpha_{ba}(t+\beta\tau_s))\langle\cos\{(\boldsymbol{k}_{bae}-\boldsymbol{k}_{ba}+\boldsymbol{k}'_{cb}-\boldsymbol{k}_{cb})\cdot\boldsymbol{r}\}\rangle_{\mathrm{av}}]\sin\omega_{ba}t,\end{aligned} \tag{41a}$$

and

$$\begin{aligned}\langle \boldsymbol{p}_{cbk_{cbe}}\rangle = {} & \sqrt{2}P_{cb}N[-B_{2\tau_s}B'_{2\tau_s}G(\alpha_{cb}(t-2\tau_s))\langle\cos\{(\boldsymbol{k}_{cbe}-2\boldsymbol{k}'_{cb}+\boldsymbol{k}_{cb})\cdot\boldsymbol{r}\}\rangle_{\mathrm{av}} \\ & + B_{(2+\beta^{-1})\tau_s}B'_{(2+\beta^{-1})\tau_s}G(\alpha_{cb}\{t-(2+\beta^{-1})\tau_s\})\langle\cos\{(\boldsymbol{k}_{cbe}-2\boldsymbol{k}'_{cb}+\boldsymbol{k}_{cb} \\ & -\boldsymbol{k}'_{ba}+\boldsymbol{k}_{ba})\cdot\boldsymbol{r}\}\rangle_{\mathrm{av}} + B_{(1+\beta^{-1})\tau_s}B'_{(1+\beta^{-1})\tau_s}G(\alpha_{cb}\{t-(1+\beta^{-1})\tau_s\}) \\ & \times\langle\cos\{(\boldsymbol{k}_{cbe}-\boldsymbol{k}'_{cb}-\boldsymbol{k}'_{ba}+\boldsymbol{k}_{ba})\cdot\boldsymbol{r}\}\rangle_{\mathrm{av}} + B_{(1-\beta^{-1})\tau_s}B'_{(1-\beta^{-1})\tau_s} \\ & \times G(\alpha_{cb}\{t-(1-\beta^{-1})\tau_s\})\langle\cos\{(\boldsymbol{k}_{cbe}-\boldsymbol{k}'_{cb}+\boldsymbol{k}'_{ba}-\boldsymbol{k}_{ba})\cdot\boldsymbol{r}\}\rangle_{\mathrm{av}} \\ & + B_{-\beta^{-1}\tau_s}B'_{-\beta^{-1}\tau_s}G(\alpha_{cb}(t+\beta^{-1}\tau_s))\langle\cos\{(\boldsymbol{k}_{cbe}-\boldsymbol{k}_{cb}+\boldsymbol{k}'_{ba}-\boldsymbol{k}_{ba})\cdot\boldsymbol{r}\}\rangle_{\mathrm{av}}] \\ & \times\sin\omega_{cb}t,\end{aligned} \tag{41b}$$

where $\boldsymbol{k}_{bae}$ and $\boldsymbol{k}_{cbe}$ are the wavevectors of the photon echoes.

From equations (41*a*) and (41*b*), we can find that the echo radiation is generated coherently in the particular direction. While the propagation directions for the normal echoes at $t = 2\tau_s$ agree with the results for the two-level system, the wavevectors for the anomalous echoes are determined by the four wavevectors at two different frequencies through the coherent coupling effect associated inherently with the resonant three-level system.

In a similar way, we can also analyse the propagation direction of photon echoes at the sum frequency ω_{ca}. What is evident from table 2 is that the complete phase matching can be realized for special combinations of the wavevectors even if the exciting pulses are non-parallel; this is found for the first time in our analysis on the three-level system.

Table 2. Summary of the propagation direction of doubly resonant photon echoes in a three-level system with the frequencies ω_{ba}, ω_{cb} and ω_{ca}. The propagation directions of the anomalous echoes depend upon the wavevectors of the exciting optical pulses with frequency different from that of the echo

Frequency of echo	Time when echo arises	Direction of echo propagation†
ω_{ba}	$2\tau_s$	$2\boldsymbol{k}'_{ab}-\boldsymbol{k}_{ab}$
	$(2+\beta)\tau_s$	$2\boldsymbol{k}'_{ab}-\boldsymbol{k}_{ab}+\boldsymbol{k}'_{bc}-\boldsymbol{k}_{bc}$
	$(1+\beta)\tau_s$	$\boldsymbol{k}'_{ab}+\boldsymbol{k}_{bc}-\boldsymbol{k}'_{bc}$
	$(1-\beta)\tau_s$	$\boldsymbol{k}'_{ab}+\boldsymbol{k}'_{bc}-\boldsymbol{k}_{bc}$
	$-\beta\tau_s$	$\boldsymbol{k}_{ab}+\boldsymbol{k}'_{bc}-\boldsymbol{k}_{bc}$
ω_{cb}	$2\tau_s$	$2\boldsymbol{k}'_{bc}-\boldsymbol{k}_{bc}$
	$(2+\beta^{-1})\tau_s$	$2\boldsymbol{k}'_{bc}-\boldsymbol{k}'_{bc}+\boldsymbol{k}'_{ab}-\boldsymbol{k}_{ab}$
	$(1+\beta^{-1})\tau_s$	$\boldsymbol{k}'_{bc}+\boldsymbol{k}_{ab}-\boldsymbol{k}'_{ab}$
	$(1-\beta^{-1})\tau_s$	$\boldsymbol{k}'_{bc}+\boldsymbol{k}'_{ab}-\boldsymbol{k}_{ab}$
	$-\beta^{-1}\tau_s$	$\boldsymbol{k}_{bc}+\boldsymbol{k}'_{ab}-\boldsymbol{k}_{ab}$
ω_{ca}	$2\tau_s$	$2\boldsymbol{k}'_{ab}-\boldsymbol{k}_{ab}+2\boldsymbol{k}'_{bc}-\boldsymbol{k}_{bc}$
	$\{1+(1+\beta)^{-1}\}\tau_s$	$2\boldsymbol{k}'_{ab}-\boldsymbol{k}_{ba}+\boldsymbol{k}'_{bc}$
	$\{1+(1+\beta^{-1})^{-1}\}\tau_s$	$2\boldsymbol{k}'_{bc}-\boldsymbol{k}_{bc}+\boldsymbol{k}'_{ab}$
	$(1+\beta)^{-1}\tau_s$	$\boldsymbol{k}'_{ab}+\boldsymbol{k}_{bc}$
	$(1+\beta^{-1})^{-1}\tau_s$	$\boldsymbol{k}_{ab}+\boldsymbol{k}'_{bc}$

‡ Subscripts *ab* and *bc* should read *ba* and *cb* respectively in entries under this heading.

7. Discussion and conclusion

The results in the previous section that the anomalous echoes are produced at times different from $2\tau_s$ is puzzling at first, since it can not be understood by the well known explanation that the second excitation pulse reverses the dephasing process so that the system rephases, at the same rate at which it dephased, to emit an intense burst of light called the photon echo. This remarkable result for the three-level system can be interpreted as follows; the transitions from $|a\rangle$ to $|b\rangle$ and from $|b\rangle$ to $|c\rangle$ (or transitions from $|b\rangle$ to $|a\rangle$ and from $|c\rangle$ to $|b\rangle$) correlate quantum mechanically with each other since the raising operators R_{ba+} and R_{cb+} (or lowering operators R_{ba-} and R_{cb-}) do not commute, so that the unitary operator $\exp\{i(\theta R_{ba1}+\phi R_{cb1})\}$ in equation (22) causes the transitions between $|a\rangle$ and $|c\rangle$ having no connection with the electric dipole-moment operator for the transition *a–c*.

For example, we consider how the echo at frequency ω_{ba} appearing at $t=(2+\beta)\tau_s$ is formed. This echo is derived from the third term in equation (30*a*) and the first term in equation (30*b*), which indicates the following process. The first two pulses induce the matrix element $\langle c|\rho|a\rangle$ from $\langle a|\rho|a\rangle$ and this element changes its phase at the rate $\Delta\omega_{ba}+\Delta\omega_{cb}$. Then, the second pulses transform this into another matrix element $\langle a|\rho|b\rangle$, which changes its phase at rate $-\Delta\omega_{ba}$. This situation directly leads to the fact that the dephasing rate is $1+\beta$ times the rephasing rate, so that the echo arises at $(2+\beta)\tau_s$

after the first pulses. Other anomalous echoes can be also explained by similar considerations.

In conclusion, we have made the theoretical analysis on the photon echo characteristics associated with the resonant three-level system by taking into account the inhomogeneous broadening of the spectral lines. Novel and interesting features are found quantitatively with respect to the echo formation time, intensity, polarization together with the propagation direction. They are expected to be examined by the well arranged experiment. These results also seem to shed light on new applications in the field of optical information processing as well as spectroscopy in the time domain.

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