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Photon echoes in a resonant three-level system with arbitrary level degeneracy

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Abstract. As a sequel to the previous paper, in which photon echoes in a resonant three-level system where the energy levels are not degenerate or have two-fold degeneracy were investigated, we report in this paper the theoretical analysis on photon echoes in a resonant multi-level system, with level degeneracy, by a general consideration. With respect to the system possessing more than three-fold degeneracy, the intensity of photon echoes depends not only on the areas of the exciting optical pulses, but also on their polarization characteristics. In particular, it was found that some anomalous echoes change their intensities with the polarization directions of exciting optical pulses which have different frequency from that of the echoes because of the coherent coupling of resonant transitions with common energy level and the interference effect of transitions through the degenerate sublevels. In the first half of this paper the general formulation on the multi-level system with arbitrary degeneracy is performed, and in the second half, the application of the general theory to some typical examples is also given.

1. Introduction

At the present time there is considerable interest in coherent nonlinear optical phenomena where optical pulses with different frequencies are resonant to the multi-energy level system, since different transitions with a common energy level combine coherently. With respect to the three-level system, Hartmann (1968) suggested the possibility of the Raman echo for the first time, and recently some novel results concerning the self-induced transparency for the two-photon resonant case (Tanno *et al* 1972a, b) and the doubly resonant photon echoes (Aihara and Inaba 1973) were reported.

In particular the coherent nonlinear effect associated with a doubly resonant interaction should exhibit characteristic phenomena inherent to the three-level system, because all the energy levels couple strongly with the radiation. These phenomena are different from the Raman or two-photon resonant type of interaction where the population of the intermediate level is extremely small due to the off-resonant character.

From this point, we presented the theoretical analysis on the doubly resonant photon echoes in a three-level system in a previous paper (Aihara and Inaba 1973, to be referred to as I). It was pointed out that the echoes arise at anomalous times, other than at the normal time expected from the simple analysis with the two-level system, depending upon the correlation between the inhomogeneous broadening of the different spectral lines. The intensity, polarization and propagation direction of the echoes were found to depend upon those of the exciting pulses which had frequency different from that of the echo.

An important problem concerning the influence of level degeneracy on the echo characteristics, however, has been left unsolved in paper I. This problem becomes essential if we consider the coherent nonlinear optical phenomena occurring in a time shorter than the homogeneous relaxation times. In this case, as the probability of the incoherent transitions among the degenerate sublevels caused by the collision in gases or the interaction with phonons in solids is negligibly small, the coherent transitions among the individual sublevels must be considered exactly.

The purpose of the present paper is to investigate photon echoes associated with the resonant interaction with a multi-level system possessing level degeneracy using general considerations. Use is made of a density operator method which has an advantage when dealing with the mixed quantum state, since the initial state is in thermodynamical equilibrium and the definite phase correlation between degenerate sublevels does not exist. In order to get a simple physical insight into the problem, we transform the density operator in the Schrödinger picture to the intermediate picture corresponding to the rotating frame representation in the classical treatment. A general formulation is derived in § 2 by introducing a unitary operator which diagonalizes the interaction hamiltonian, and by obtaining the exact time evolution operator without using the perturbation approach. In § 3, we apply our general results to some typical examples to make our detailed discussion more concrete.

2. General formulation for the formation of photon echoes

We consider the system composed of many particles possessing multi-energy levels with arbitrary degeneracy which interacts resonantly with optical waves with different frequencies. When dealing with this system, we must make use of the density operator for the whole system, since photon echoes are phenomena inherent in the many particle system. However, if the interaction between particles does not exist, the Liouville equation for the whole system can be separated into the equations for the individual particles, and the density operator for the whole system can be obtained as the direct product of those of the individual particles. Therefore, the starting point of our analysis is the solution of the one-particle problem.

The hamiltonian for a particle interacting resonantly with the radiation field with different frequencies can be written, in general, as

$$H = H_0 + H', \quad (1)$$

$$H_0 = \sum_{\mu} \mathcal{E}_{\mu} \sum_{m_{\mu}} |\alpha_{\mu} j_{\mu} m_{\mu}\rangle \langle \alpha_{\mu} j_{\mu} m_{\mu}|, \quad (2)$$

$$\begin{aligned} H' &= \sum_{\mu, \nu} (-\mathbf{p}_{\mu\nu}) \cdot \mathbf{E}_{\mu\nu}(t) \\ &= \sum_{\mu, \nu} (-\sqrt{2}) E_{\mu\nu 0}(t) (\mathbf{p}_{\mu\nu} \cdot \mathbf{u}_{\mu\nu}) \cos \omega_{\mu\nu} t, \end{aligned} \quad (3)$$

where \mathcal{E}_{μ} is the energy eigenvalue of the free hamiltonian H_0 , $\mathbf{p}_{\mu\nu}$ is the electric dipole moment operator between levels μ and ν , $\mathbf{E}_{\mu\nu}(t)$ is the electric field vector resonant to the transition between levels μ and ν , and $E_{\mu\nu 0}(t)$ and $\mathbf{u}_{\mu\nu}$ are its envelope function and polarization vector, respectively.

Taking the direction perpendicular to the electric field vector as the quantization axis and using the Wigner–Eckart theorem†, equation (3) becomes through the expression in terms of the basis vectors for the standard representation

$$H' = - \sum_{\mu, \nu} \sqrt{2} P_{\mu\nu} E_{\mu\nu 0}(t) \sum_{m_\mu, m_\nu} (\chi_{m_\mu m_\nu}^{j_\mu j_\nu} |\alpha_\mu j_\mu m_\mu\rangle \langle \alpha_\nu j_\nu m_\nu| + \chi_{m_\nu m_\mu}^{j_\nu j_\mu} |\alpha_\nu j_\nu m_\nu\rangle \langle \alpha_\mu j_\mu m_\mu|) \times (\mathbf{e}_+ \cdot \mathbf{u}_{\mu\nu}) \cos \omega_{\mu\nu} t + \text{hermitian adjoint}, \tag{4}$$

where $P_{\mu\nu}$ is the reduced matrix element, \mathbf{e}_+ is the circular polarization vector, and $\chi_{m_\alpha m_\beta}^{j_\alpha j_\beta}$ is given by $\chi_{m_\alpha m_\beta}^{j_\alpha j_\beta} = (2j_\alpha + 1)^{-1/2} |j_\beta 1 m_\beta 1\rangle \langle j_\alpha m_\alpha|$.

Considering the time evolution of the system, it is convenient to use the density operator which is applicable to the mixed state as well as the pure state. The density operator $\sigma(t)$ satisfies the Liouville equation

$$i\hbar \frac{d\sigma(t)}{dt} = [H, \sigma(t)]. \tag{5}$$

We now transform the density operator $\sigma(t)$ in the Schrödinger picture to the intermediate picture, as follows:

$$\rho(t) = e^{iSt} \sigma(t) e^{-iSt} \tag{6}$$

where S is a hermitian operator defined by

$$S = \sum_{\lambda} S_{\lambda} \sum_{m_{\lambda}} |\alpha_{\lambda} j_{\lambda} m_{\lambda}\rangle \langle \alpha_{\lambda} j_{\lambda} m_{\lambda}| \tag{7}$$

and eigenvalues S_{λ} are determined to satisfy the relation $S_{\mu} - S_{\nu} = \omega_{\mu\nu}$. This unitary transformation leads to the equation of motion for $\rho(t)$

$$i\hbar \frac{d\rho(t)}{dt} = [\hbar\Delta + H'_s, \rho(t)]. \tag{8}$$

where

$$H'_s = e^{iSt} H' e^{-iSt}, \tag{9}$$

$$\Delta = \hbar^{-1} H_0 - S. \tag{10}$$

Using the rotating wave approximation, equation (9) becomes

$$H'_s = - \sum_{\mu, \nu} \frac{1}{\sqrt{2}} P_{\mu\nu} E_{\mu\nu 0}(t) \sum_{m_\mu, m_\nu} (\chi_{m_\mu m_\nu}^{j_\mu j_\nu} |\mu m_\mu\rangle \langle \nu m_\nu| + \chi_{m_\nu m_\mu}^{j_\nu j_\mu} |\nu m_\nu\rangle \langle \mu m_\mu|) (\mathbf{e}_+ \cdot \mathbf{u}_{\mu\nu}) + \text{hermitian adjoint}, \tag{11}$$

where the simplified notation $|\lambda m_\lambda\rangle$ is used instead of $|\alpha_\lambda j_\lambda m_\lambda\rangle$.

No matter what we consider the resonant interaction, we must take into account the off-resonance caused by the inhomogeneous broadening of the energy level, so that the difference between the eigenvalues of Δ defined by equation (10), $\Delta\omega_{\mu\nu}$, takes the non-zero value determined by

$$\Delta\omega_{\mu\nu} = (\hbar^{-1} E_{\mu} - S_{\mu}) - (\hbar^{-1} E_{\nu} - S_{\nu}) = \hbar^{-1} (E_{\mu} - E_{\nu}) - (S_{\mu} - S_{\nu}) = \Omega_{\mu\nu} - \omega_{\mu\nu}. \tag{12}$$

We are now in a position to solve the case where the system is irradiated by a sequence of two simultaneous optical pulses with different frequencies which coincide with the

† See for example Messiah A 1962 *Quantum Mechanics* vol 2 (Amsterdam: North-Holland).

centre frequencies of the inhomogeneously broadened spectral lines of the system. For sufficiently intense exciting pulses, the magnitude of the off-resonance can be considered to be much smaller than the interaction energy, and then we ignore Δ for the duration of the pulse to obtain

$$i\hbar \frac{d\rho(t)}{dt} = [H'_s, \rho(t)]. \quad (13)$$

For the period of absence of the pulses, we have

$$i\hbar \frac{d\rho(t)}{dt} = [\Delta, \rho(t)]. \quad (14)$$

In the case where the envelope function differs from another one corresponding to the different frequency only by a multiplicative number independent of time, equation (13) can be solved exactly. For the optical pulses lasting from t_0 to $t_0 + \tau$, the formal solution is expressed by

$$\rho(t_0 + \tau) = \exp\left(-i\hbar^{-1} \int_{t_0}^{t_0 + \tau} dt H'_s(t)\right) \rho(t_0) \exp\left(i\hbar^{-1} \int_{t_0}^{t_0 + \tau} dt H'_s(t)\right). \quad (15)$$

In order to obtain the matrix element of $\rho(t_0 + \tau)$, we introduce the unitary operator V defined by

$$V^\dagger \int_{t_0}^{t_0 + \tau} H'_s(t) dt V = D, \quad (16)$$

where D is a diagonal operator in the standard representation. Then equation (15) can be rewritten as

$$\rho(t_0 + \tau) = T \rho(t_0) T^\dagger \quad (17)$$

where T is the time evolution operator given by

$$T = \exp(-iVDV^\dagger) = V \exp(-iD)V^\dagger. \quad (18)$$

On the other hand, equation (14) can be solved simply as

$$\rho(t) = \exp\{-i\Delta(t-t')\} \rho(t') \exp\{i\Delta(t-t')\}. \quad (19)$$

Using the solutions (17) and (19), we derive the expression for the density operator after a sequence of two optical pulses

$$\rho(t) = \exp\{-i\Delta(t-\tau_s)\} T' \exp(-i\Delta\tau_s) T \rho(0) T^\dagger \exp(i\Delta\tau_s) T'^{\dagger} \exp\{i\Delta(t-\tau_s)\}, \quad (20)$$

where the prime denotes the operator for the second pulse and τ_s is a time interval between two optical pulses.

We suppose that the many particle system is initially in thermodynamical equilibrium, and the energy separations between the ground state and the excited states are much larger than the thermal energy. Thus we have the initial density operator given by

$$\rho(0) = (2j_a + 1)^{-1} \sum_{m_a} |am_a\rangle \langle am_a|, \quad (21)$$

where label a denotes the ground state. Substituting this equation into equation (20),

we obtain the expression for the matrix element of $\rho(t)$ in terms of the matrix element of T as

$$\begin{aligned} \langle \mu m_\mu | \rho(t) | \nu m_\nu \rangle &= (2j_a + 1)^{-1} \sum_{m_a} \langle \mu m_\mu | \exp\{-i\Delta(t - \tau_s)\} T' \exp(-i\Delta\tau_s) T | a m_a \rangle \\ &\quad \times \langle \nu m_\nu | \exp\{-i\Delta(t - \tau_s)\} T' \exp(-i\Delta\tau_s) T | a m_a \rangle^* \\ &= (2j_a + 1)^{-1} \sum_{\xi, \eta} \exp\{-i\Delta\omega_{\mu\nu}t + i(\Delta\omega_{\mu\nu} + \Delta\omega_{\eta\xi})\tau_s\} \\ &\quad \times \sum_{m_a, m_\xi, m_\eta} \langle \mu m_\mu | T' | \xi m_\xi \rangle \langle \xi m_\xi | T | a m_a \rangle \langle \nu m_\nu | T' | \eta m_\eta \rangle^* \langle \eta m_\eta | T | a m_a \rangle^*. \end{aligned} \quad (22)$$

In order to study the various characteristics of photon echoes, we need an induced electric dipole moment by calculating the expectation values of the electric dipole moment operator, as follows:

$$\langle \mathbf{p}(t) \rangle = \text{Tr}[\sigma(t)\mathbf{p}] = \sum_{\mu, \nu} \text{Tr}[\sigma(t)\mathbf{p}_{\mu\nu}] = \sum_{\mu, \nu} \langle \mathbf{p}_{\mu\nu}(t) \rangle \quad (23)$$

and

$$\begin{aligned} \langle \mathbf{p}_{\mu\nu}(t) \rangle &= \sum_{\xi, \eta, m_\xi, m_\eta} \langle \xi m_\xi | e^{-iS^\dagger} \rho(t) e^{iS} | \eta m_\eta \rangle \langle \eta m_\eta | \mathbf{p}_{\mu\nu} | \xi m_\xi \rangle \\ &= P_{\mu\nu} \sum_{m_\mu, m_\nu} \{ \chi_{m_\nu, m_\mu}^{j_\nu j_\mu} \langle \nu m_\nu | \rho(t) | \mu m_\mu \rangle \exp(i\omega_{\mu\nu}t) \\ &\quad + \chi_{m_\nu, m_\mu}^{j_\nu j_\mu} \langle \mu m_\mu | \rho(t) | \nu m_\nu \rangle \exp(-i\omega_{\mu\nu}t) \} e_+ + \text{CC} \\ &= P_{\mu\nu} \sum_{m_\mu, m_\nu} (\chi_{m_\nu, m_\mu}^{j_\nu j_\mu} e_+ - \chi_{m_\mu, m_\nu}^{j_\mu j_\nu} e_-) \langle \mu m_\mu | \rho(t) | \nu m_\nu \rangle \exp(-i\omega_{\mu\nu}t) + \text{CC}. \end{aligned} \quad (24)$$

Substituting equation (22) into equation (24), we obtain a final expression for an induced electric dipole moment with frequency $\omega_{\mu\nu}$ after a sequence of optical pulses

$$\begin{aligned} \langle \mathbf{p}_{\mu\nu}(t) \rangle &= (2j_a + 1)^{-1} P_{\mu\nu} \sum_{\xi, \eta} \exp\{-i\Delta\omega_{\mu\nu}t + i(\Delta\omega_{\mu\nu} + \Delta\omega_{\eta\xi})\tau_s\} \sum_{m_\mu, m_\nu} (\chi_{m_\nu, m_\mu}^{j_\nu j_\mu} e_+ - \chi_{m_\mu, m_\nu}^{j_\mu j_\nu} e_-) \\ &\quad \times \sum_{m_a, m_\xi, m_\eta} \langle \mu m_\mu | T' | \xi m_\xi \rangle \langle \xi m_\xi | T | a m_a \rangle \\ &\quad \times \langle \nu m_\nu | T' | \eta m_\eta \rangle^* \langle \eta m_\eta | T | a m_a \rangle^* \exp(-i\omega_{\mu\nu}t) + \text{CC}. \end{aligned} \quad (25)$$

In the case where the frequencies of the incident optical pulses are at the centre frequencies of the inhomogeneously broadened spectral lines, $\Delta\omega_{\mu\nu}$ and $\Delta\omega_{\eta\xi}$ can be written

$$\begin{aligned} \Delta\omega_{\mu\nu} &= \alpha_{\mu\nu}x, \\ \Delta\omega_{\eta\xi} &= \alpha_{\eta\xi}x. \end{aligned} \quad (26)$$

Here x is a physical parameter causing the inhomogeneous broadening of the energy levels, and we neglect the higher order terms in x with good approximation. For example, in gases, x is the velocity of a particle along the line of sight, while, in solids, it is the deviation of the crystalline field from its mean value. In order to obtain the essential characteristics of the radiation emitted spontaneously from the inhomogeneously broadened system, we must multiply the distribution function for x in equation (25), and integrate

over x . Thus we obtain

$$\begin{aligned}
 \langle p_{\mu\nu}(t) \rangle &= N(2j_a + 1)^{-1} P_{\mu\nu} \sum_{\xi, \eta} G \{ \alpha_{\mu\nu} t - (\alpha_{\mu\nu} + \alpha_{\eta\xi}) \tau_s \} \sum_{m_\mu, m_\nu} (\chi_{m_\nu m_\mu}^{j_\mu j_\nu} e_+ - \chi_{m_\mu m_\nu}^{j_\mu j_\nu} e_-) \\
 &\times \sum_{m_a, m_\xi, m_\mu} \langle \mu m_\mu | T | \xi m_\xi \rangle \langle \xi m_\xi | T | a m_a \rangle \\
 &\times \langle \nu m_\nu | T | \eta m_\eta \rangle^* \langle \eta m_\eta | T | a m_a \rangle^* \exp(-i\omega_{\mu\nu} t) + \text{CC}
 \end{aligned} \tag{27}$$

where N is the number of particles and G is the Fourier transform of the distribution function for x .

From equation (27), we can understand the remarkable result that photon echoes are also produced at anomalous times different from $2\tau_s$, depending upon the values $\alpha_{\eta\xi}/\alpha_{\mu\nu}$. This novel result is caused by the fact that the matrix element of the density operator after a sequence of exciting optical pulses, $\langle \mu m_\mu | \rho | \nu m_\nu \rangle$, arises from other matrix elements before the second exciting pulse, $\langle \xi m_\xi | \rho | \eta m_\eta \rangle$, as indicated in equation (22), so that the rate of the dephasing process after the first pulses is different from that of the inphasing process after the second pulses as suggested in equation (25). One should remark that the operators T and T' can have the nonvanishing matrix elements between the levels for which the interaction hamiltonian does not have the matrix element, because of the coupling of transitions between the levels through a common energy level.

When the Doppler effect gives rise to the inhomogeneous broadening of lines as in gases, the times of the anomalous echoes are determined by the ratio of the energy separations since the relation $\alpha_{\eta\xi}/\alpha_{\mu\nu} = \Omega_{\eta\xi}/\Omega_{\mu\nu}$ holds. However, for the ions in a ligand field where the spatial fluctuation of the static crystalline field contributes to the inhomogeneous broadening of the 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 also the configuration interaction. Consequently, photon echoes are produced at times different from the case of gases even if the ratio of the energy separations $\Omega_{\eta\xi}/\Omega_{\mu\nu}$ is the same.

3. Evaluation of echo characteristics

In this section we try to apply the general result obtained in the previous section to some typical examples to derive explicit expressions and to discuss them in detail.

3.1. The case $j_a = j_b = j_c = \frac{1}{2}$

As we have considered this case in detail in paper I, we mention only the essential point. When each energy level has double degeneracy, the problem can be reduced to a non-degenerate one by dividing the whole Hilbert space into two subspaces between which the interaction hamiltonian does not have the matrix element. This results in the intensity and polarization characteristics becoming independent of each other. However, one should note that the coupling of the transitions through a common energy level leads to the characteristics of the photon echoes, such as intensity, polarization and propagation direction, depending upon those of the exciting pulses which have different frequencies from that of the echo.

3.2. The case $j_a = j_c = 1, j_b = 0$

Since we have selected the quantization axis perpendicular to the direction of the exciting optical pulses, five states take part in the problem, as shown in figure 1. In this case, the

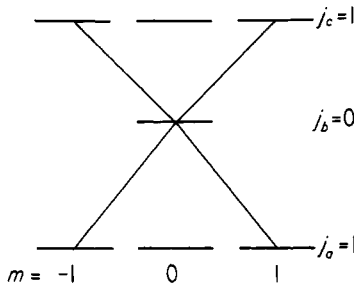


Figure 1. Schematic representation of the three-level system for $j_a = j_c = 1$ and $j_b = 0$. Full lines connect the states between which the interaction hamiltonian has a nonzero matrix element when the quantization axis is selected along the direction perpendicular to that of the exciting electric field.

interaction hamiltonian H'_s expressed generally in equation (11) becomes

$$\begin{aligned}
 H'_s = & -\frac{1}{2\sqrt{3}}P_{ba}E_{ba0}(t)\{|3\rangle\langle 1| \exp(-i\delta_{ba}) + |3\rangle\langle 2| \exp(i\delta_{ba})\} \\
 & -\frac{1}{2\sqrt{3}}P_{cb}E_{cb0}(t)\{|5\rangle\langle 3| \exp(-i\delta_{cb}) + |4\rangle\langle 3| \exp(i\delta_{cb})\} \\
 & + \text{hermitian adjoint,}
 \end{aligned} \tag{28}$$

where the simplified notations $|1\rangle = |a-1\rangle, |2\rangle = |a\rangle, |3\rangle = |b0\rangle, |4\rangle = |c-1\rangle$ and $|5\rangle = |c1\rangle$ are used. Introducing the pulse area defined by

$$\theta_{\mu\nu} = \left(\frac{2}{3}\right)^{1/2}P_{\mu\nu} \int_{t_0}^{t_0+\tau} E_{\mu\nu 0}(t) dt, \tag{29}$$

we obtain the matrix element of T defined by equation (18) as follows;

$$\langle 1|T|1\rangle = \langle 2|T|2\rangle = 1 - \frac{1}{2} \cos^2 \gamma (1 - \cos \frac{1}{2}\psi), \tag{30a}$$

$$\langle 3|T|3\rangle = \cos \frac{1}{2}\psi, \tag{30b}$$

$$\langle 4|T|4\rangle = \langle 5|T|5\rangle = 1 - \frac{1}{2} \sin^2 \gamma (1 - \cos \frac{1}{2}\psi), \tag{30c}$$

$$\langle 3|T|1\rangle = -\langle 3|T|2\rangle^* = -i\sqrt{\frac{1}{2}} \cos \gamma \sin \frac{1}{2}\psi \exp(-i\delta_{ba}), \tag{30d}$$

$$\langle 5|T|3\rangle = -\langle 4|T|3\rangle^* = -i\sqrt{\frac{1}{2}} \sin \gamma \sin \frac{1}{2}\psi \exp(-i\delta_{cb}), \tag{30e}$$

$$\langle 5|T|1\rangle = \langle 4|T|2\rangle^* = -\frac{1}{2} \sin \gamma \cos \gamma (1 - \cos \frac{1}{2}\psi) \exp\{-i(\delta_{cb} + \delta_{ba})\}, \tag{30f}$$

$$\langle 4|T|1\rangle = \langle 5|T|2\rangle^* = -\frac{1}{2} \sin \gamma \cos \gamma (1 - \cos \frac{1}{2}\psi) \exp\{i(\delta_{cb} - \delta_{ba})\}, \tag{30g}$$

$$\langle 2|T|1\rangle = -\frac{1}{2} \cos^2 \gamma (1 - \cos \frac{1}{2}\psi) \exp(-2i\delta_{ba}), \tag{30h}$$

$$\langle 5|T|4\rangle = -\frac{1}{2} \sin^2 \gamma (1 - \cos \frac{1}{2}\psi) \exp(-2i\delta_{cb}), \tag{30i}$$

where $\delta_{\mu\nu}$ is an angle between the direction of the electric field of the exciting pulse with

frequency $\omega_{\mu\nu}$ and the reference axis, and $\gamma = \tan^{-1}(\theta_{cb}/\theta_{ba})$ and $\psi = (\theta_{ba}^2 + \theta_{cb}^2)^{1/2}$. Other matrix elements can easily be obtained from the unitarity of T , that is, $\langle f|T|i\rangle$ differs from $\langle i|T|f\rangle$ only by a sign of δ . The matrix element of T , $\langle f|T|i\rangle$, means physically the probability amplitude that if a particle is initially in a state $|i\rangle$, then after a pulse the particle will be in a state $|f\rangle$. One should pay attention to their phases as well as their absolute values. For example, the transition probabilities from $|1\rangle$ to $|3\rangle$ and from $|2\rangle$ to $|3\rangle$ are the same, but the phases of their transition probability amplitudes differ corresponding to the polarization direction of the exciting pulse, as indicated in equation (30d).

Substituting equations (30a)–(30i) into equation (27), we obtain the induced polarization, which is given in the appendix. The results concerning the echo polarization and the dependence of the echo intensity on the polarizations of the exciting pulses in this case are summarized in table 1. The principal results obtained are: (a) in the case where the polarization directions of the sequence of two exciting pulses with the same frequency are parallel, the intensity characteristics are the same as the nondegenerate case except for a multiplicative constant; (b) the echoes, except for those appearing at $-\beta\tau_s$ and $-\beta^{-1}\tau_s$ ($\beta = \alpha_{cb}/\alpha_{ba}$), are polarized in the direction of that for the second pulse with the same frequency; (c) the echo intensity appearing at the time $2\tau_s$ varies as $\cos^2(\delta'_{\mu\nu} - \delta_{\mu\nu})$ in the same way as the two-level system (Gordon *et al* 1969), while the anomalous echo intensities, except for two echoes, also or only depend upon the polarization directions of the exciting pulses, with different frequencies from that of the echo, as $\cos^2(\delta'_{\mu\nu} - \delta_{\mu\nu})$; (d) with respect to the echoes appearing at the times $-\beta\tau_s$ and $-\beta^{-1}\tau_s$, both the echo polarization and the change of the echo intensity with the polarization directions of the exciting pulses depend upon the areas of the exciting pulses through the matrix elements of T' for the nondegenerate case, $\langle a|T'|a\rangle$ or $\langle c|T'|c\rangle$.

The result (c) inherent to the three-level system is caused by the two facts that, first, the transition probability between the ground state $|am_a\rangle$ and the higher excited state $|cm_c\rangle$ takes a nonzero value caused by the coupling of the transitions $b-a$ and $c-b$, and, second, some matrix elements of the density operator related to the different degenerate states after the first exciting pulse interfere with each other when forming the final density operator by the second exciting pulse. The result (d) is also substantial to the three-level

Table 1. Summary of echo polarizations and dependence of echo intensities on the polarizations of exciting optical pulses, for $j_a = j_c = 1$ and $j_b = 0$. The analytical expressions of δ_1 , Q_1 , δ_2 and Q_2 include the areas for the second exciting pulses θ'_{ba} and θ'_{cb} , and hence for ψ' and γ' , and are given in the appendix. $\beta = \alpha_{cb}/\alpha_{ba}$.

Echo frequency	Echo formation time	Echo polarization	Polarization dependence of echo intensity
ω_{ba}	$2\tau_s$	δ'_{ba}	$\cos^2(\delta'_{ba} - \delta_{ba})$
	$(1 + \beta)\tau_s$	δ'_{ba}	$\cos^2(\delta'_{cb} - \delta_{cb})$
	$(2 + \beta)\tau_s$	δ'_{ba}	$\cos^2(\delta'_{ba} - \delta_{ba}) \cos^2(\delta'_{cb} - \delta_{cb})$
	$(1 - \beta)\tau_s$	δ'_{ba}	$\cos^2(\delta'_{cb} - \delta_{cb})$
	$-\beta\tau_s$	δ_1	Q_1^2
ω_{cb}	$2\tau_s$	δ'_{cb}	$\cos^2(\delta'_{cb} - \delta_{cb})$
	$(1 + \beta^{-1})\tau_s$	δ'_{cb}	$\cos^2(\delta'_{ba} - \delta_{ba})$
	$(2 + \beta^{-1})\tau_s$	δ'_{cb}	$\cos^2(\delta'_{cb} - \delta_{cb}) \cos^2(\delta'_{ba} - \delta_{ba})$
	$(1 - \beta^{-1})\tau_s$	δ'_{cb}	$\cos^2(\delta'_{ba} - \delta_{ba})$
	$-\beta^{-1}\tau_s$	δ_2	Q_2^2

system. This phenomenon is caused by the fact that these two anomalous echoes are formed through the matrix elements of T' between the degenerate sublevels.

3.3. The case $j_a = j_c = 0, j_b = 1$

This case (figure 2) is more complicated than the two examples studied previously because the eigenvalues of the interaction hamiltonian depend upon the polarizations of the exciting pulses, so that the magnitude of the matrix elements of T as well as their phases takes a cumbersome form. Therefore, we show only the results in table 2, for the case where the polarization direction of the exciting pulse with the different frequency is parallel. What is evident from table 2 is that the polarization and dependence of the intensity appearing at $(1 + \beta)\tau_s$ and $(1 + \beta^{-1})\tau_s$ on the polarizations of the exciting pulses, change with the pulse area of the second pulse. Furthermore, with respect to this example, the intensity of echoes appearing at $(2 + \beta)\tau_s$ and $(2 + \beta^{-1})\tau_s$ is independent of the polarization of the exciting pulses. These results can be understood in analogy with the case studied in § 3.1, since the matrix element of the density operator between the non-degenerate ground and higher excited states after the first pulse, contributes to such echoes.

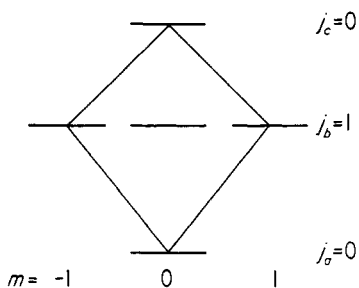


Figure 2. Schematic representation of the three-level system for $j_a = j_c = 0$ and $j_b = 1$. Full lines connect the states between which the interaction hamiltonian has a nonzero matrix element when the quantization axis is selected along the direction perpendicular to that of the exciting electric field.

Table 2. Summary of echo polarizations and dependence of echo intensities on the polarizations of exciting optical pulses, for $j_a = j_c = 0$ and $j_b = 1$. Analytical expressions of δ_3 and Q_3 include ψ' , and are given in the appendix. $\beta = \alpha_{cb}/\alpha_{ba}$.

Echo frequency	Echo formation time	Echo polarization	Polarization dependence of echo intensity
ω_{ba}	$2\tau_s$	δ'	$\cos^2(\delta' - \delta)$
	$(1 + \beta)\tau_s$	δ_3	Q_3^2
	$(2 + \beta)\tau_s$	δ'	constant
	$(1 - \beta)\tau_s$	δ'	$\cos^2(\delta' - \delta)$
	$-\beta\tau_s$	δ'	constant
ω_{cb}	$2\tau_s$	δ'	$\cos^2(\delta' - \delta)$
	$(1 + \beta^{-1})\tau_s$	δ_3	Q_3^2
	$(2 + \beta^{-1})\tau_s$	δ'	constant
	$(1 - \beta^{-1})\tau_s$	δ'	$\cos^2(\delta' - \delta)$
	$-\beta^{-1}\tau_s$	δ'	constant

3.4. The case $j_a = j_b = j_c = 1$

In this case, the nine-dimensional Hilbert space is composed of two subspaces between which the interaction hamiltonian does not have the matrix element as shown in figure 3. Therefore, the problem can be reduced to the cases studied in §§ 3.2 and 3.3, and the results are shown in table 3.

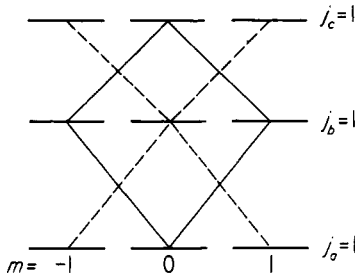


Figure 3. Schematic representation of the three-level system for $j_a = j_b = j_c = 1$. Full and broken lines show the two independent sets of transitions caused by the optical wave with linear polarization perpendicular to the quantization axis.

Table 3. Summary of echo polarization and dependence of echo intensity on the polarizations of exciting optical pulses, for $j_a = j_b = j_c = 1$. Analytical expressions of $\delta_4, Q_4, \delta_5, Q_5, \delta_6,$ and Q_6 include the areas of the second exciting pulses θ'_{ba} and θ'_{cb} and hence for ψ' and γ' , and are given in the appendix. $\beta = \alpha_{cb}/\alpha_{ba}$.

Echo frequency	Echo formation time	Echo polarization	Polarization dependence of echo intensity
ω_{ba}	$2\tau_s$	δ'	$\cos^2(\delta' - \delta)$
	$(1 + \beta)\tau_s$	δ_4	Q_4^2
	$(2 + \beta)\tau_s$	δ'	$\{1 + \cos(\delta' - \delta)\}^2$
	$(1 - \beta)\tau_s$	δ'	$\cos^2(\delta' - \delta)$
	$-\beta\tau_s$	δ_5	Q_5^2
ω_{cb}	$2\tau_s$	δ'	$\cos^2(\delta' - \delta)$
	$(1 + \beta^{-1})\tau_s$	δ_4	Q_4^2
	$(2 + \beta^{-1})\tau_s$	δ'	$\{1 + \cos(\delta' - \delta)\}^2$
	$(1 - \beta^{-1})\tau_s$	δ'	$\cos^2(\delta' - \delta)$
	$-\beta^{-1}\tau_s$	δ_6	Q_6^2

3.5. The case $j_a = j_c = \frac{3}{2}, j_b = \frac{1}{2}$

A distinctive feature of this case is that the magnitudes of the matrix element of the electric dipole moment operator between the different degenerate sublevels are different because of the nature of the Clebsch–Gordan coefficient. Therefore, the interference effect between the transitions through the different degenerate sublevels does not cause the echoes to vanish completely even when the polarization direction of the second exciting pulse is perpendicular to that of the first exciting pulse. Hence the echo intensity varies as $1 - k \sin^2(\delta'_{\mu\nu} - \delta_{\mu\nu})$ ($k = \frac{3}{4}$ or $\frac{1-5}{16}$) and the polarization direction of the echo radiation is not parallel to that of the second exciting pulse as shown in table 4.

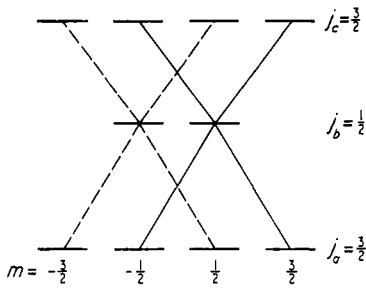


Figure 4. Schematic representation of the three-level system for $j_a = j_c = \frac{3}{2}$ and $j_b = \frac{1}{2}$. Full and broken lines show the two independent sets of transitions caused by the optical waves with linear polarization perpendicular to the quantization axis.

Table 4. Summary of echo polarization and dependence of echo intensities on the polarizations of exciting optical pulses, for $j_a = j_c = \frac{3}{2}$ and $j_b = \frac{1}{2}$. Analytical expressions of δ_7 , Q_7 , δ_8 and Q_8 include the areas for the second exciting pulses θ'_{ba} and θ'_{cb} and hence for ψ' and γ' , and are given in the appendix. $\beta = \alpha_{cb}/\alpha_{ba}$.

Echo frequency	Echo formation time	Echo polarization	Polarization dependence of echo intensity
ω_{ba}	$2\tau_s$	$\delta'_{ba} + \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba})$	$1 - \frac{1}{6} \sin^2(\delta'_{ba} - \delta_{ba})$
	$(1 + \beta)\tau_s$	$\delta'_{ba} - \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb})$	$1 - \frac{1}{6} \sin^2(\delta'_{cb} - \delta_{cb})$
	$(2 + \beta)\tau_s$	$\delta'_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba}) - \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb})$	$\{1 - \frac{3}{4} \sin^2(\delta'_{ba} - \delta_{ba})\} \{1 - \frac{1}{6} \sin^2(\delta'_{cb} - \delta_{cb})\}$
	$(1 - \beta)\tau_s$	$\delta'_{ba} + \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb})$	$1 - \frac{1}{6} \sin^2(\delta'_{cb} - \delta_{cb})$
	$-\beta\tau_s$	δ_7	Q_7^2
ω_{cb}	$2\tau_s$	$\delta'_{cb} + \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb})$	$1 - \frac{1}{6} \sin^2(\delta'_{cb} - \delta_{cb})$
	$(1 + \beta^{-1})\tau_s$	$\delta'_{cb} - \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba})$	$1 - \frac{1}{6} \sin^2(\delta'_{ba} - \delta_{ba})$
	$(2 + \beta^{-1})\tau_s$	$\delta'_{cb} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb}) - \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba})$	$\{1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb})\} \{1 - \frac{1}{6} \sin^2(\delta'_{ba} - \delta_{ba})\}$
	$(1 - \beta^{-1})\tau_s$	$\delta'_{cb} + \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba})$	$1 - \frac{1}{6} \sin^2(\delta'_{ba} - \delta_{ba})$
	$-\beta^{-1}\tau_s$	δ_8	Q_8^2

3.6. The case $j_a = j_b = \frac{1}{2}$, $j_c = \frac{3}{2}$

This case is very complicated compared with the earlier examples since we must analyse the coupling of the Q and R branch transitions, while in the cases studied in §§ 3.1–3.5. the two transitions are of the same type of branch. This fact leads to the cumbersome expression for the photon echo arising at time $(2 + \beta^{-1})\tau_s$, as shown in table 5. Table 5 also shows that the normal echoes with frequencies ω_{ba} and ω_{cb} have the same characteristics as the cases studied in §§ 3.1 and 3.5, respectively.

In the case of $j_a = \frac{3}{2}$ and $j_b = j_c = \frac{1}{2}$, we obtain the results by exchanging the subscripts cb and ba , and β and β^{-1} , respectively.

Before completing the description of this section, one should note that the mathematical expressions in the last columns of tables 1–5 only show the dependence of echo

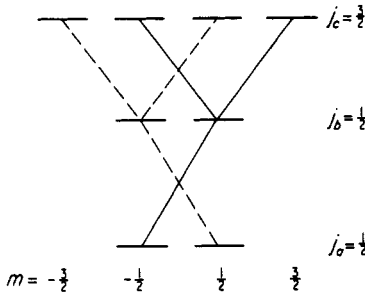


Figure 5. Schematic representation of the three-level system for $j_a = \frac{1}{2}$ and $j_b = j_c = \frac{3}{2}$. Full and broken lines show the two independent sets of transitions caused by the optical wave with linear polarization perpendicular to the quantization axis.

Table 5. Summary of echo polarizations and dependence of echo intensities on the polarizations of exciting optical pulses for $j_a = \frac{1}{2}$ and $j_b = j_c = \frac{3}{2}$. Analytical expressions of δ_g and Q_g include the areas for the second exciting pulses θ'_{ba} and θ'_{cb} , and hence for ψ' and γ' , and are given in the appendix. $\beta = \alpha_{cb}/\alpha_{ba}$.

Echo frequency	Echo formation time	Echo polarization	Polarization dependence of echo intensity
ω_{ba}	$2\tau_s$	$2\delta'_{ba} - \delta_{ba}$	constant
	$(1 + \beta)\tau_s$	$\delta'_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})$	$1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb})$
	$(2 + \beta)\tau_s$	$2\delta'_{ba} - \delta_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})$	$1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb})$
	$(1 - \beta)\tau_s$	$\delta'_{ba} - \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})$	$1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb})$
	$-\beta\tau_s$	$\delta'_{ba} - \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})$	$1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb})$
ω_{cb}	$2\tau_s$	$\delta'_{cb} + \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb})$	$1 - \frac{5}{16} \sin^2(\delta'_{cb} - \delta_{cb})$
	$(1 + \beta^{-1})\tau_s$	$\delta'_{cb} + \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba})$	$1 - \frac{3}{4} \sin^2(\delta'_{ba} - \delta_{ba})$
	$(2 + \beta^{-1})\tau_s$	$\delta'_{cb} + \tan^{-1} \frac{1}{2} \tan\{\delta'_{ba} - \delta_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})\}$	$\{1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb})\} [1 - \frac{3}{4} \sin^2\{\delta'_{ba} - \delta_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})\}]$
	$(1 - \beta^{-1})\tau_s$	$\delta'_{cb} - \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba})$	$1 - \frac{3}{4} \sin^2(\delta'_{ba} - \delta_{ba})$
	$-\beta^{-1}\tau_s$	δ_g	Q_g^2

intensity on the polarizations of the exciting pulses and do not indicate the dependence on the areas of the exciting pulses even for the expressions involving the pulse areas. The full expressions for the electric polarization which gives rise to photon echoes are summarized in the appendix for some typical examples treated in this section. It is to be remarked that, when the polarization directions of the exciting pulses are all parallel, the intensity characteristics coincide with those for the nondegenerate case with respect to all the examples analysed in this section.

The results obtained in this section are also applicable to the case of $\mathcal{E}_b > \mathcal{E}_c$, that is, the resonance Raman type of interaction, only by exchanging the sign of β in tables 1–5. In the case of $\mathcal{E}_a > \mathcal{E}_b$, that is, the doubly resonant interaction with the common ground state, the dependence of echo intensity on the areas for the first pulses is different because of the difference of the initial conditions.

4. Conclusion

In the present paper, we have made a theoretical study of photon echoes associated with a multi-level system by taking into account the correlation between the inhomogeneous broadenings of the different spectral lines. In the first half of this paper the general formulation applicable to the many-particle system with arbitrary degeneracy was derived, and in the second half, the application of the general theory to some important examples was given. Novel and interesting features were obtained quantitatively with respect to the polarization and intensity characteristics of the echo radiation. In particular, it is possible for the anomalous echoes to provide extensive information about the energy level structure, kinetics of the interaction between particles, and the atomic coherence inherent in the multi-level system. The results obtained in this paper seem to indicate that experimental studies on nonlinear optical behaviour in coherently excited many-particle systems would be worthwhile.

Appendix. The analytical expressions for the electric polarization which give rise to the echoes

A.1. The case $j_a = j_b = j_c = \frac{1}{2}$

For this case we refer the reader to paper I.

A.2. The case $j_a = j_c = 1, j_b = 0$

$$\begin{aligned}
 \langle \mathbf{p}(t) \rangle_{\text{echo}} = & \frac{2NP_{ba}}{3\sqrt{3}} [-A_{2\tau_s} A'_{2\tau_s} \cos(\delta'_{ba} - \delta_{ba}) G\{\alpha_{ba}(t - 2\tau_s)\} (\mathbf{i} \cos \delta'_{ba} + \mathbf{j} \sin \delta'_{ba}) \\
 & + A_{(1+\beta)\tau_s} A'_{(1+\beta)\tau_s} \cos(\delta'_{cb} - \delta_{cb}) G\{\alpha_{ba}(t - (1+\beta)\tau_s)\} (\mathbf{i} \cos \delta'_{ba} + \mathbf{j} \sin \delta'_{ba}) \\
 & + A_{(2+\beta)\tau_s} A'_{(2+\beta)\tau_s} \cos(\delta'_{ba} - \delta_{ba}) \cos(\delta'_{cb} - \delta_{cb}) \\
 & \times G\{\alpha_{ba}(t - (2+\beta)\tau_s)\} (\mathbf{i} \cos \delta'_{ba} + \mathbf{j} \sin \delta'_{ba}) \\
 & + A_{(1-\beta)\tau_s} A'_{(1-\beta)\tau_s} \cos(\delta'_{cb} - \delta_{cb}) G\{\alpha_{ba}(t - (1-\beta)\tau_s)\} (\mathbf{i} \cos \delta'_{ba} + \mathbf{j} \sin \delta'_{ba}) \\
 & - A_{-\beta\tau_s} \sin \gamma' \sin \frac{1}{2}\psi' Q_1 G\{\alpha_{ba}(t + \beta\tau_s)\} (\mathbf{i} \cos \delta_1 + \mathbf{j} \sin \delta_1)] \sin \omega_{ba} t \\
 & + \frac{2NP_{cb}}{3\sqrt{3}} [-B_{2\tau_s} B'_{2\tau_s} \cos(\delta'_{cb} - \delta_{cb}) G\{\alpha_{cb}(t - 2\tau_s)\} (\mathbf{i} \cos \delta'_{cb} + \mathbf{j} \sin \delta'_{cb}) \\
 & + B_{(1+\beta^{-1})\tau_s} B'_{(1+\beta^{-1})\tau_s} \cos(\delta'_{ba} - \delta_{ba}) \\
 & \times G\{\alpha_{cb}(t - (1+\beta^{-1})\tau_s)\} (\mathbf{i} \cos \delta'_{cb} + \mathbf{j} \sin \delta'_{cb}) \\
 & + B_{(2+\beta^{-1})\tau_s} B'_{(2+\beta^{-1})\tau_s} \cos(\delta'_{ba} - \delta_{ba}) \cos(\delta'_{cb} - \delta_{cb}) \\
 & \times G\{\alpha_{cb}(t - (2+\beta^{-1})\tau_s)\} (\mathbf{i} \cos \delta'_{cb} + \mathbf{j} \sin \delta'_{cb}) \\
 & + B_{(1-\beta^{-1})\tau_s} B'_{(1-\beta^{-1})\tau_s} \cos(\delta'_{ba} - \delta_{ba}) \\
 & \times G\{\alpha_{cb}(t - (1-\beta^{-1})\tau_s)\} (\mathbf{i} \cos \delta'_{cb} + \mathbf{j} \sin \delta'_{cb}) \\
 & - B_{-\beta^{-1}\tau_s} \cos \gamma' \sin \frac{1}{2}\psi' Q_2 G\{\alpha_{cb}(t + \beta^{-1}\tau_s)\} (\mathbf{i} \cos \delta_2 + \mathbf{j} \sin \delta_2)] \sin \omega_{cb} t.
 \end{aligned}
 \tag{A.1}$$

Here

$$A_{2\tau_s} = \cos \gamma \sin \frac{1}{2}\psi (\cos^2 \gamma \cos \frac{1}{2}\psi + \sin^2 \gamma), \quad (\text{A.2})$$

$$A'_{2\tau_s} = \cos^2 \gamma' \sin^2 \frac{1}{2}\psi', \quad (\text{A.3})$$

$$A_{(2+\beta)\tau_s} = \frac{1}{2} \sin 2\gamma (1 - \cos \frac{1}{2}\psi) (\cos^2 \gamma \cos \frac{1}{2}\psi + \sin^2 \gamma), \quad (\text{A.4})$$

$$A'_{(2+\beta)\tau_s} = \frac{1}{2} \sin 2\gamma' \cos \gamma' \sin \frac{1}{2}\psi' (1 - \cos \frac{1}{2}\psi'), \quad (\text{A.5})$$

$$A_{(1+\beta)\tau_s} = \frac{1}{2} \sin 2\gamma \cos \gamma \sin \frac{1}{2}\psi (1 - \cos \frac{1}{2}\psi), \quad (\text{A.6})$$

$$A'_{(1+\beta)\tau_s} = \frac{1}{2} \sin 2\gamma' \cos \frac{1}{2}\psi' (1 - \cos \frac{1}{2}\psi'), \quad (\text{A.7})$$

$$A_{(1-\beta)\tau_s} = A_{(1+\beta)\tau_s}, \quad (\text{A.8})$$

$$A'_{(1-\beta)\tau_s} = \frac{1}{2} \sin 2\gamma' \sin^2 \frac{1}{2}\psi', \quad (\text{A.9})$$

$$A_{-\beta\tau_s} = A_{(2+\beta)\tau_s}, \quad (\text{A.10})$$

$$A'_{-\beta\tau_s} = \sin \gamma' \sin \frac{1}{2}\psi' (\cos^2 \gamma' \cos \frac{1}{2}\psi' + \sin^2 \gamma'), \quad (\text{A.11})$$

$$B_{2\tau_s} = A_{(1+\beta)\tau_s}, \quad (\text{A.12})$$

$$B'_{2\tau_s} = \sin^2 \gamma' \sin^2 \frac{1}{2}\psi', \quad (\text{A.13})$$

$$B_{(2+\beta^{-1})\tau_s} = A_{(2+\beta)\tau_s}, \quad (\text{A.14})$$

$$B'_{(2+\beta^{-1})\tau_s} = \frac{1}{2} \sin 2\gamma' \sin \frac{1}{2}\psi' (1 - \cos \frac{1}{2}\psi'), \quad (\text{A.15})$$

$$B_{(1+\beta^{-1})\tau_s} = A_{2\tau_s}, \quad (\text{A.16})$$

$$B'_{(1+\beta^{-1})\tau_s} = A'_{(1+\beta)\tau_s}, \quad (\text{A.17})$$

$$B_{(1-\beta^{-1})\tau_s} = A_{2\tau_s}, \quad (\text{A.18})$$

$$B'_{(1-\beta^{-1})\tau_s} = A'_{(1-\beta)\tau_s}, \quad (\text{A.19})$$

$$B_{-\beta^{-1}\tau_s} = A_{(2+\beta)\tau_s}, \quad (\text{A.20})$$

$$B'_{-\beta^{-1}\tau_s} = \cos \gamma' \sin \frac{1}{2}\psi' (\cos^2 \gamma' + \sin^2 \gamma' \cos \frac{1}{2}\psi'), \quad (\text{A.21})$$

$$\delta_1 = \delta'_{ba} - \tan^{-1} \frac{\tan(\delta'_{ba} - \delta_{ba})}{\langle a|T'|a \rangle}, \quad (\text{A.22})$$

$$Q_1 = \cos(\delta'_{cb} - \delta_{cb}) \{ \langle a|T'|a \rangle^2 \cos^2(\delta'_{ba} - \delta_{ba}) + \sin^2(\delta'_{ba} - \delta_{ba}) \}^{1/2}, \quad (\text{A.23})$$

$$\delta_2 = \delta'_{cb} - \tan^{-1} \frac{\tan(\delta'_{cb} - \delta_{cb})}{\langle c|T'|c \rangle}, \quad (\text{A.24})$$

$$Q_2 = \cos(\delta'_{ba} - \delta_{ba}) \{ \langle c|T'|c \rangle^2 \cos^2(\delta'_{cb} - \delta_{cb}) + \sin^2(\delta'_{cb} - \delta_{cb}) \}^{1/2}, \quad (\text{A.25})$$

where

$$\langle a|T'|a \rangle = \cos^2 \gamma' \cos \frac{1}{2}\psi' + \sin^2 \gamma',$$

$$\langle c|T'|c \rangle = \sin^2 \gamma' \cos \frac{1}{2}\psi' + \cos^2 \gamma'.$$

A.3. The case $j_a = j_c = 0, j_b = 1$

$$\begin{aligned}
 \langle \rho(t) \rangle_{\text{echo}} = & \frac{2NP_{ba}}{\sqrt{3}} [-A_{2\tau_s} A'_{2\tau_s} \cos(\delta' - \delta) G\{\alpha_{ba}(t - 2\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta')] \\
 & + \frac{1}{2} A_{(1+\beta)\tau_s} \sin 2\gamma' (1 - \cos \frac{1}{2}\psi') Q_3 G\{\alpha_{ba}(t - (1 + \beta)\tau_s)\} (\mathbf{i} \cos \delta_3 + \mathbf{j} \sin \delta_3) \\
 & + A_{(2+\beta)\tau_s} A'_{(2+\beta)\tau_s} G\{\alpha_{ba}(t - (2 + \beta)\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta') \\
 & + A_{(1-\beta)\tau_s} A'_{(1-\beta)\tau_s} \cos(\delta' - \delta) G\{\alpha_{ba}(t - (1 - \beta)\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta') \\
 & - A_{-\beta\tau_s} A'_{-\beta\tau_s} G\{\alpha_{ba}(t + \beta\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta')] \sin \omega_{ba} t \\
 & + \frac{2NP_{cb}}{\sqrt{3}} [-B_{2\tau_s} B'_{2\tau_s} \cos(\delta' - \delta) G\{\alpha_{cb}(t - 2\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta')] \\
 & + \frac{1}{2} B_{(1+\beta^{-1})\tau_s} \sin 2\gamma' (1 - \cos \frac{1}{2}\psi') Q_3 G\{\alpha_{cb}(t - (1 + \beta^{-1})\tau_s)\} (\mathbf{i} \cos \delta_3 + \mathbf{j} \sin \delta_3) \\
 & + B_{(2+\beta^{-1})\tau_s} B'_{(2+\beta^{-1})\tau_s} G\{\alpha_{cb}(t - (2 + \beta^{-1})\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta') \\
 & + B_{(1-\beta^{-1})\tau_s} B'_{(1-\beta^{-1})\tau_s} \cos(\delta' - \delta) G\{\alpha_{cb}(t - (1 - \beta^{-1})\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta') \\
 & - B_{-\beta^{-1}\tau_s} B'_{-\beta^{-1}\tau_s} G\{\alpha_{cb}(t + \beta^{-1}\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta')] \sin \omega_{cb} t, \tag{A.26}
 \end{aligned}$$

where

$$\delta_3 = \delta' - \tan^{-1} \frac{\tan(\delta' - \delta)}{\langle b|T'|b \rangle}, \tag{A.27}$$

$$Q_3 = \{ \langle b|T'|b \rangle^2 \cos^2(\delta' - \delta) + \sin^2(\delta' - \delta) \}^{-1/2}, \tag{A.28}$$

and

$$\langle b|T'|b \rangle = \cos \frac{1}{2}\psi'.$$

A.4. The case $j_a = j_b = j_c = 1$

$$\begin{aligned}
 \langle \rho(t) \rangle_{\text{echo}} = & \frac{2NP_{ba}}{3\sqrt{6}} [-2A_{2\tau_s} A'_{2\tau_s} \cos(\delta' - \delta) G\{\alpha_{ba}(t - 2\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta')] \\
 & + \frac{1}{2} A_{(1+\beta)\tau_s} \sin 2\gamma' (1 - \cos \frac{1}{2}\psi') Q_4 G\{\alpha_{ba}(t - (1 + \beta)\tau_s)\} (\mathbf{i} \cos \delta_4 + \mathbf{j} \sin \delta_4) \\
 & + A_{(2+\beta)\tau_s} A'_{(2+\beta)\tau_s} \{1 + \cos(\delta' - \delta)\} G\{\alpha_{ba}(t - (2 + \beta)\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta') \\
 & + 2A_{(1-\beta)\tau_s} A'_{(1-\beta)\tau_s} \cos(\delta' - \delta) G\{\alpha_{ba}(t - (1 - \beta)\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta') \\
 & - A_{-\beta\tau_s} \sin \gamma' \sin \frac{1}{2}\psi' Q_5 G\{\alpha_{ba}(t + \beta\tau_s)\} (\mathbf{i} \cos \delta_5 + \mathbf{j} \sin \delta_5) \sin \omega_{ba} t \\
 & + \frac{4NP_{cb}}{3\sqrt{6}} [-2B_{2\tau_s} B'_{2\tau_s} \cos(\delta' - \delta) G\{\alpha_{cb}(t - 2\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta')] \\
 & + \frac{1}{2} B_{(1+\beta^{-1})\tau_s} \sin 2\gamma' (1 - \cos \frac{1}{2}\psi') Q_4 G\{\alpha_{cb}(t - (1 + \beta^{-1})\tau_s)\} (\mathbf{i} \cos \delta_4 + \mathbf{j} \sin \delta_4) \\
 & + B_{(2+\beta^{-1})\tau_s} B'_{(2+\beta^{-1})\tau_s} \{1 + \cos(\delta' - \delta)\} \\
 & \times G\{\alpha_{cb}(t - (2 + \beta^{-1})\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta') \\
 & + 2B_{(1-\beta^{-1})\tau_s} B'_{(1-\beta^{-1})\tau_s} \cos(\delta' - \delta) G\{\alpha_{cb}(t - (1 - \beta^{-1})\tau_s)\} (\mathbf{i} \cos \delta' + \mathbf{j} \sin \delta') \\
 & - B_{-\beta^{-1}\tau_s} \cos \gamma' \sin \frac{1}{2}\psi' Q_6 G\{\alpha_{cb}(t + \beta^{-1}\tau_s)\} (\mathbf{i} \cos \delta_6 + \mathbf{j} \sin \delta_6) \sin \omega_{cb} t. \tag{A.29}
 \end{aligned}$$

Here

$$\delta_4 = \delta' - \tan^{-1} \frac{\tan(\delta' - \delta)}{2\langle b|T|b\rangle}, \quad (\text{A.30})$$

$$Q_4 = \{4\langle b|T|b\rangle^2 \cos^2(\delta' - \delta) + \sin^2(\delta' - \delta)\}^{1/2}, \quad (\text{A.31})$$

$$\delta_5 = \delta' - \tan^{-1} \frac{\tan(\delta' - \delta)}{2\langle a|T|a\rangle}, \quad (\text{A.32})$$

$$Q_5 = \{4\langle a|T|a\rangle^2 \cos^2(\delta' - \delta) + \sin^2(\delta' - \delta)\}^{1/2}, \quad (\text{A.33})$$

$$\delta_6 = \delta' - \tan^{-1} \frac{\tan(\delta' - \delta)}{2\langle c|T|c\rangle}, \quad (\text{A.34})$$

$$Q_6 = \{4\langle c|T|c\rangle^2 \cos^2(\delta' - \delta) + \sin^2(\delta' - \delta)\}^{1/2}. \quad (\text{A.35})$$

A.5. The case $j_a = j_c = \frac{3}{2}$, $j_b = \frac{1}{2}$

$$\begin{aligned} \langle \mathbf{p}(t) \rangle_{\text{echo}} = & \frac{NP_{ba}}{\sqrt{6}} [-A_{2\tau_s} A'_{2\tau_s} \{1 - \frac{1}{6} \sin^2(\delta'_{ba} - \delta_{ba})\}^{1/2} G\{\alpha_{ba}(t - 2\tau_s)\} \\ & \times \{i \cos(\delta'_{ba} + \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba})) + j \sin(\delta'_{ba} + \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba}))\} \\ & + A_{(1+\beta)\tau_s} A'_{(1+\beta)\tau_s} \{1 - \frac{1}{6} \sin^2(\delta'_{cb} - \delta_{cb})\}^{1/2} G\{\alpha_{ba}(t - (1+\beta)\tau_s)\} \\ & \times \{i \cos(\delta'_{ba} - \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb})) + j \sin(\delta'_{ba} - \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb}))\} \\ & + A_{(2+\beta)\tau_s} A'_{(2+\beta)\tau_s} \{1 - \frac{3}{4} \sin^2(\delta'_{ba} - \delta_{ba})\}^{1/2} \{1 - \frac{1}{6} \sin^2(\delta'_{cb} - \delta_{cb})\}^{1/2} \\ & \times G\{\alpha_{ba}(t - (2+\beta)\tau_s)\} \{i \cos(\delta'_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba}) \\ & - \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb})) + j \sin(\delta'_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba}) \\ & - \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb}))\} + A_{(1-\beta)\tau_s} A'_{(1-\beta)\tau_s} \{1 - \frac{1}{6} \sin^2(\delta'_{cb} - \delta_{cb})\}^{1/2} \\ & \times G\{\alpha_{ba}(t - (1-\beta)\tau_s)\} \{i \cos(\delta'_{ba} + \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb})) \\ & + j \sin(\delta'_{ba} + \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb}))\} \\ & - A_{-\beta\tau_s} \sin \gamma' \sin \frac{1}{2} \psi' Q_7 G\{\alpha_{ba}(t + \beta\tau_s)\} (i \cos \delta_7 + j \sin \delta_7) \sin \omega_{ba} t \\ & + \frac{NP_{ba}}{\sqrt{6}} [-B_{2\tau_s} B'_{2\tau_s} \{1 - \frac{1}{6} \sin^2(\delta'_{cb} - \delta_{cb})\}^{1/2} G\{\alpha_{cb}(t - 2\tau_s)\} \\ & \times \{i \cos(\delta'_{cb} + \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb})) + j \sin(\delta'_{cb} + \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb}))\} \\ & + B_{(1+\beta^{-1})\tau_s} B'_{(1+\beta^{-1})\tau_s} \{1 - \frac{1}{6} \sin^2(\delta'_{ba} - \delta_{ba})\}^{1/2} G\{\alpha_{cb}(t - (1+\beta^{-1})\tau_s)\} \\ & \times \{i \cos(\delta'_{cb} - \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba})) \\ & + j \sin(\delta'_{cb} - \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba}))\} \\ & + B_{(2+\beta^{-1})\tau_s} B'_{(2+\beta^{-1})\tau_s} \{1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb})\}^{1/2} \\ & \times \{1 - \frac{1}{6} \sin^2(\delta'_{ba} - \delta_{ba})\}^{1/2} G\{\alpha_{cb}(t - (2+\beta^{-1})\tau_s)\} \\ & \times \{i \cos(\delta'_{cb} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb}) - \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba})) \\ & + j \sin(\delta'_{cb} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb}) - \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba}))\} \end{aligned}$$

$$\begin{aligned}
 & + B_{(1-\beta^{-1})\tau_s} B'_{(1-\beta^{-1})\tau_s} \left\{ 1 - \frac{15}{16} \sin^2(\delta'_{ba} - \delta_{ba}) \right\}^{1/2} \\
 & \times G\{\alpha_{cb}(t - (1 - \beta^{-1})\tau_s)\} \{ i \cos(\delta'_{cb} + \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba})) \\
 & + j \sin(\delta'_{cb} + \tan^{-1} \frac{1}{4} \tan(\delta'_{ba} - \delta_{ba})) \} \\
 & - B_{-\beta^{-1}\tau_s} \cos \gamma' \sin \frac{1}{2} \psi' Q_8 G\{\alpha_{cb}(t + \beta^{-1}\tau_s)\} (i \cos \delta_8 + j \sin \delta_8) \sin \omega_{cb} t.
 \end{aligned} \tag{A.36}$$

Here

$$\delta_7 = \delta'_{ba} + \tan^{-1} \frac{3r_{ba+} \sin \phi_{ba+} - r_{ba-} \sin \phi_{ba-}}{3r_{ba+} \cos \phi_{ba+} + r_{ba-} \cos \phi_{ba-}}, \tag{A.37}$$

$$\begin{aligned}
 Q_7 = \{ & 1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb}) \}^{1/2} \{ (\frac{3}{4}r_{ba+} \cos \phi_{ba+} + \frac{1}{4}r_{ba-} \cos \phi_{ba-})^2 \\
 & + (\frac{3}{4}r_{ba+} \sin \phi_{ba+} - \frac{1}{4}r_{ba-} \sin \phi_{ba-})^2 \}^{1/2}
 \end{aligned} \tag{A.38}$$

$$\delta_8 = \delta'_{cb} + \tan^{-1} \frac{3r_{cb+} \sin \phi_{cb+} - r_{cb-} \sin \phi_{cb-}}{3r_{cb+} \cos \phi_{cb+} + r_{cb-} \cos \phi_{cb-}}, \tag{A.39}$$

$$\begin{aligned}
 Q_8 = \{ & 1 - \frac{3}{4} \sin^2(\delta'_{ba} - \delta_{ba}) \}^{1/2} \{ (\frac{3}{4}r_{cb+} \cos \phi_{cb+} + \frac{1}{4}r_{cb-} \cos \phi_{cb-})^2 \\
 & + (\frac{3}{4}r_{cb+} \sin \phi_{cb+} - \frac{1}{4}r_{cb-} \sin \phi_{cb-})^2 \}^{1/2}.
 \end{aligned} \tag{A.40}$$

In these formulae, the parameters are

$$r_{ba\pm} = \{ \langle a|T'|a \rangle^2 \cos^2(\delta'_{ba} - \delta_{ba}) + \frac{1}{4}(3 \pm \langle a|T'|a \rangle)^2 \sin^2(\delta'_{ba} - \delta_{ba}) \}^{1/2} \tag{A.41}$$

$$\phi_{ba\pm} = \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb}) - \tan^{-1} \left(\frac{3 \pm \langle a|T'|a \rangle}{2 \langle a|T'|a \rangle} \tan(\delta'_{ba} - \delta_{ba}) \right), \tag{A.42}$$

$$r_{cb\pm} = \{ \langle c|T'|c \rangle^2 \cos^2(\delta'_{cb} - \delta_{cb}) + \frac{1}{4}(3 \pm \langle c|T'|c \rangle)^2 \sin^2(\delta'_{cb} - \delta_{cb}) \}^{1/2} \tag{A.43}$$

$$\phi_{cb\pm} = \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba}) - \tan^{-1} \left(\frac{3 \pm \langle c|T'|c \rangle}{2 \langle c|T'|c \rangle} \tan(\delta'_{cb} - \delta_{cb}) \right). \tag{A.44}$$

A.6. The case $j_a = j_b = \frac{1}{2}, j_c = \frac{3}{2}$

$$\begin{aligned}
 \langle \mathbf{p}(t) \rangle_{\text{echo}} = & \sqrt{\frac{2}{3}} NP_{ba} [-A_{2\tau_s} A'_{2\tau_s} G\{\alpha_{ba}(t - 2\tau_s)\} \{ i \cos(2\delta'_{ba} - \delta_{ba}) + j \sin(2\delta'_{ba} - \delta_{ba}) \} \\
 & + A_{(1+\beta)\tau_s} A'_{(1+\beta)\tau_s} \{ 1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb}) \}^{1/2} G\{\alpha_{ba}(t - (1 + \beta)\tau_s)\} \\
 & \times \{ i \cos(\delta'_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})) + j \sin(\delta'_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})) \} \\
 & + A_{(2+\beta)\tau_s} A'_{(2+\beta)\tau_s} \{ 1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb}) \}^{1/2} G\{\alpha_{ba}(t - (2 + \beta)\tau_s)\} \\
 & \times \{ i \cos(2\delta'_{ba} - \delta_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})) \\
 & + j \sin(2\delta'_{ba} - \delta_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})) \} \\
 & + A_{(1-\beta)\tau_s} A'_{(1-\beta)\tau_s} \{ 1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb}) \}^{1/2} G\{\alpha_{ba}(t - (1 - \beta)\tau_s)\} \\
 & \times \{ i \cos(\delta'_{ba} - \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})) + j \sin(\delta'_{ba} - \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})) \} \\
 & - A_{-\beta\tau_s} A'_{-\beta\tau_s} \{ 1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb}) \}^{1/2} G\{\alpha_{ba}(t + \beta\tau_s)\} \\
 & \times \{ i \cos(\delta'_{ba} - \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})) \\
 & + j \sin(\delta'_{ba} - \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})) \} \sin \omega_{ba} t \\
 & + \sqrt{\frac{2}{3}} NP_{cb} [-B_{2\tau_s} B'_{2\tau_s} \{ 1 - \frac{15}{16} \sin^2(\delta'_{cb} - \delta_{cb}) \}^{1/2} G\{\alpha_{cb}(t - 2\tau_s)\}
 \end{aligned}$$

$$\begin{aligned}
 & \times \{i \cos(\delta'_{cb} + \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb})) + j \sin(\delta'_{cb} + \tan^{-1} \frac{1}{4} \tan(\delta'_{cb} - \delta_{cb}))\} \\
 & + B_{(1+\beta^{-1})\tau_s} B'_{(1+\beta^{-1})\tau_s} \{1 - \frac{3}{4} \sin^2(\delta'_{ba} - \delta_{ba})\}^{1/2} G\{\alpha_{cb}(t - (1 + \beta^{-1})\tau_s)\} \\
 & \times \{i \cos(\delta'_{cb} + \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba})) + j \sin(\delta'_{cb} + \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba}))\} \\
 & + B_{(2+\beta^{-1})\tau_s} B'_{(2+\beta^{-1})\tau_s} \{1 - \frac{3}{4} \sin^2(\delta'_{cb} - \delta_{cb})\}^{1/2} \\
 & \times \{1 - \frac{3}{4} \sin^2(\delta'_{ba} - \delta_{ba} + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb}))\}^{1/2} \\
 & \times G\{\alpha_{cb}(t - (2 + \beta^{-1})\tau_s)\} \{i \cos(\delta'_{cb} + \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba} \\
 & + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb})) + j \sin(\delta'_{cb} + \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba} \\
 & + \tan^{-1} \frac{1}{2} \tan(\delta'_{cb} - \delta_{cb}))\} \\
 & + B_{(1-\beta^{-1})\tau_s} B'_{(1-\beta^{-1})\tau_s} \{1 - \frac{3}{4} \sin^2(\delta'_{ba} - \delta_{ba})\}^{1/2} G\{\alpha_{cb}(t - (1 - \beta^{-1})\tau_s)\} \\
 & \times \{i \cos(\delta'_{cb} - \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba})) + j \sin(\delta'_{cb} - \tan^{-1} \frac{1}{2} \tan(\delta'_{ba} - \delta_{ba}))\} \\
 & - B_{-\beta^{-1}\tau_s} \cos \gamma' \sin \frac{1}{2} \psi' Q_9 G\{\alpha_{cb}(t + \beta^{-1}\tau_s)\} \\
 & \times (i \cos \delta_9 + j \sin \delta_9) \sin \omega_{cb} t, \tag{A.45}
 \end{aligned}$$

where

$$\delta_9 = \delta'_{cb} - \tan^{-1} \frac{3r_+ \sin \phi_+ - r_- \sin \phi_-}{3r_+ \cos \phi_+ + r_- \cos \phi_-}, \tag{A.46}$$

$$Q_9 = \{(\frac{3}{4}r_+ \cos \phi_+ + \frac{1}{4}r_- \cos \phi_-)^2 + (\frac{3}{4}r_+ \sin \phi_+ - \frac{1}{4}r_- \sin \phi_-)^2\}^{1/2} \tag{A.47}$$

and

$$r_{\pm} = \{\langle c|T|c\rangle^2 \cos^2(\delta'_{cb} - \delta_{cb}) + \frac{1}{4}(1 \pm \langle c|T|c\rangle)^2 \sin^2(\delta'_{cb} - \delta_{cb})\}^{1/2}, \tag{A.48}$$

$$\phi_{\pm} = \tan^{-1} \left(\frac{1 \pm \langle c|T|c\rangle}{2\langle c|T|c\rangle} \tan(\delta'_{cb} - \delta_{cb}) \right). \tag{A.49}$$

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