Fourth-order interference on polarization beats in a four-level system

Yanpeng Zhang, Liqun Sun, and Tiantong Tang
Department of Electronic Science and Technology, Xi'an Jiaotong University, Xi'an 710049, China
Panming Fu
Institute of Physics, Chinese Academy of Sciences, Beijing 100080, China

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We have employed chaotic and phase-diffusion models to study the effects of laser fourth-order coherence on polarization beats with phase-conjugation geometry in a four-level system (PBFS). We found that the temporal behavior of the beat signal depends on the stochastic properties of the lasers and the transverse relaxation rate of the transition. The modulation terms of the beat signal depend on the second-order coherence function, which is determined by the laser line shape. Inasmuch as different stochastic models of the laser field affect only the fourth-order coherence function, they have little influence on the general temporal modulation behavior of the beat signal. The different roles of phase fluctuation and amplitude fluctuation are pointed out. The cases that pump beams have either narrow-band or broadband linewidth are considered, and it is found that for both cases a Doppler-free precision in the measurement of the energy-level difference between two states that are dipolar forbidden from the ground state can be achieved. We also discuss the difference between the PBFS and ultrafast modulation spectroscopy from a physical viewpoint. © 2000 Optical Society of America

1. INTRODUCTION

For many years, much effort has been devoted to studying the effects of laser field fluctuations on nonlinear optics. The partial-coherence properties of pump beams have been studied by Eichler et al. They formulated a second-order coherence-function theory in which the Bragg reflection signal intensity is represented as an absolute square of a nonlinear polarization that is averaged over the stochastic realizations of the electromagnetic field. A similar theory was described by Grossman and Shenwell. According to these theories, gratings cannot be induced by partially coherent light from two independent sources. Also, no Bragg reflection signal can be observed when the relative time delay between two pump beams that are generated from a single laser is much longer than the coherence time. Because it is the signal intensity that is measured, the correct procedure for treating this problem is to average the absolute square of the polarization over the random variable of the stochastic process. Using this method, Trebino et al. studied the effect of pulse width and grating decay on the formation of a thermal grating. In this paper we develop a unified theory that involves a fourth-order coherence function to study the influence of partial-coherence properties of pump beams on polarization beats. Polarization beats, which originate from the interference between macroscopic polarizations, have attracted much attention recently. They are closely related to quantum beat spectroscopy, which appears in conventional time-resolved fluorescence and in time-resolved nonlinear laser spectroscopy. DeBeer et al. performed the first ultrafast modulation spectroscopy (UMS) experiment in sodium vapor. The beat signal exhibits 1.9-ps modulation, corresponding to the sodium D-line split when the time delay between two double-frequency pump beams increases. Fu et al. then analyzed the UMS with phase-conjugate geometry in a Doppler-broadened system by a second-order coherence-function theory. Fu et al. found that a Doppler-free precision in the measurement of the energy-level splitting could be achieved.

In this paper we report the fourth-order effects of field correlation on phase-conjugation geometry in a four-level system (PBFS). We first assume that the laser sources are chaotic fields. A chaotic field, which is used to describe a multimode laser source, is characterized by the fluctuation of both the amplitude and the phase of the field. Another commonly used stochastic model is the phase-diffusion model, which is used to describe an amplitude-stabilized laser source. This model assumes that the amplitude of the laser field is a constant, whereas its phase fluctuates as random process. Using two radiation models, we studied the influence of various quantities, such as laser linewidth, transverse relaxation rate, and longitudinal relaxation rate. One of the relevant problems is stationary four-wave mixing (FWM) with incoherent light sources, which was proposed by Morita and Yajima to achieve ultrafast temporal resolution of relaxation processes. Because they assumed that the laser linewidth is much longer than the transverse relaxation rate, their theory cannot be used to study the effect of the light's bandwidth on the Bragg reflection signal. Asaka et al. considered the finite linewidth effect; however, the constant-background contribution was ignored in their analysis. Our fourth-order coherence function theory includes both the finite light bandwidth effect and the constant-background contribution. The different
roles of the phase fluctuation and the amplitude fluctuation in the time domain were pointed out. If PBFS is employed for measurement of the energy-level difference, the advantages are that the energy-level difference between states can be widely separated and a Doppler-free precision in the measurement can be achieved. We also investigated the relationship between PBFS and other Doppler-free techniques in the frequency and time domains. We found that PBFS is closely related to Doppler-free two-photon absorption spectroscopy with a resonant intermediate state and to the sum-frequency trilevel photon echo when the pump beams have narrowband and broadband linewidths, respectively. However, it possesses the main advantages of these techniques in the frequency domain and in the time domain.

2. BASIC THEORY

PBFS is a polarization beat phenomenon that originates from the interference between two two-photon processes. Let us consider a four-level system (Fig. 1) with a ground state 0, an intermediate state 1, and two excited states 2 and 3. States between 0 and 1 and between 1 and 2 (3) are coupled by dipolar transition with resonant frequencies 1 and 12 (13), respectively, whereas states between 2 and 3 and between 0 and 2 (3) are dipolar forbidden. We consider in this four-level system a double-frequency time-delay FWM experiment in which beams 2 and 3 consist of two frequency components, 2 and 3, whereas beam 1 has frequency 1 (Fig. 2). We assume that 1 1 and 3 3 ( 3) or (3) are coupled by dipolar transition with resonant frequencies 1 and 12 (13); therefore 1 and 2 (3) will drive the transitions from 0 (1) and from 1 to 2 (3), respectively. In this double-frequency time-delay FWM, beam 1 with frequency 1 and the 2 (3) frequency component of beam 2 induce coherence between 0 and 2 (3) by a two-photon transition, which is then probed by the 2 (3) frequency component of beam 3. These are two-photon FWM with a resonant intermediate state, and the frequency of the signal equals 1. We are interested in the dependence of the beat signal’s intensity on the relative time delay between 2 and 3.

The complex electric fields of beam 2, 2 , and of beam 3, , can be written as

\[ E_{p2} = e_{2}u_{2}(t)\exp[i(k_{2} \cdot r - \omega_{2}t)] + e_{3}u_{3}(t)\exp[i(k_{3} \cdot r - \omega_{3}t)], \]

\[ E_{p3} = e_{2}u_{2}(t - \tau)\exp[i(k_{2} \cdot r - \omega_{2}(t - \tau) + \omega_{2}\tau)] + e_{3}u_{3}(t - \tau)\exp[i(k_{3} \cdot r - \omega_{3}(t - \tau) + \omega_{3}\tau)], \]

where \( e_{1} \) and \( k_{1} \) (\( e_{1}' \), \( k_{1}' \)) are the constant field amplitude and the wave vector of the \( \omega_{1} \) component in beam 2 (beam 3), respectively. \( u_{1}(t) \) is a dimensionless statistical factor that contains phase and amplitude fluctuations. We assume that the 2 (3) component of 2 and 3 comes from a single laser source and that \( \tau \) is the time delay of beam 3 with respect to beam 2. Beam 1 is assumed to be quasi-monochromatic light. The complex electric fields of beam 1 can be written as

\[ E_{p1} = e_{1}\exp[i(k_{1} \cdot r - \omega_{1}t)], \]

where \( \omega_{1}, e_{1} \), and \( k_{1} \) are the frequency, the field amplitude, and the wave vector of the field, respectively.

We employ perturbation theory to calculate the density matrix elements. In the following perturbation chains:

\[ (I) \rho_{00}^{(0)} \rightarrow \rho_{10}^{(1)} \rightarrow \rho_{20}^{(2)} \rightarrow \rho_{30}^{(3)} \]

\[ (II) \rho_{00}^{(0)} \rightarrow \rho_{10}^{(1)} \rightarrow \rho_{20}^{(2)} \rightarrow \rho_{30}^{(3)} \]

we obtain the third-order off-diagonal density matrix element \( \rho_{30}^{(3)} \), which has wave vector \( k_{2} - k_{3}' + k_{1} \) or \( k_{3} - k_{2}' + k_{1} \). The nonlinear polarization \( P_{30}^{(3)} \) that is responsible for the phase-conjugate FWM signal is given by averaging over the velocity distribution function \( v_{W}(v) \). Thus \( P_{30}^{(3)} = N\mu_{1} \int_{-\infty}^{\infty} dv_{W}(v)\rho_{30}^{(3)}(v) \), where \( v \) is the atomic velocity and \( N \) is the density of atoms. For a Doppler-broadened atomic system we have \( v_{W}(v) = \frac{1}{\sqrt{2\pi}\sigma^{2}}\exp[-(v/\sigma^{2})^{2}] \).

The FWM signal is proportional to the average of the absolute square of \( P_{30}^{(3)} \) over the random variable of the stochastic process \( \langle |P_{30}^{(3)}|^{2} \rangle \), which involves fourth- and second-order coherence functions of \( u_{1}(t) \) in phase-conjugation geometry, whereas the FWM signal intensity in DeBeers’s self-diffraction geometry is related to the sixth-order coherence function of the incident fields. We first assume that beam 2 (beam 3) is multimode thermal source. \( u_{1}(t) \) has Gaussian statistics, with its fourth-order coherence function satisfying

\[ \langle u_{1}(t_{1})u_{1}^{*}(t_{2})u_{1}^{*}(t_{3})u_{1}^{*}(t_{4}) \rangle = \langle u_{1}(t_{1})u_{1}^{*}(t_{3})u_{1}^{*}(t_{2})u_{1}^{*}(t_{4}) \rangle + \langle u_{1}(t_{1})u_{1}^{*}(t_{4}) \rangle \times \langle u_{1}(t_{2})u_{1}^{*}(t_{3}) \rangle, \quad i = 2, 3. \]

(1)

Furthermore, assuming that beam 2 (beam 3) has a Lorentzian line shape, we have

\[ \langle u_{1}(t_{1})u_{1}^{*}(t_{2}) \rangle = \exp(-\alpha_{1}|t_{1} - t_{2}|), \quad i = 2, 3. \]

(2)
where $\alpha_i = \frac{1}{2} \delta \omega_i$, with $\delta \omega_i$ the linewidth of the laser with frequency $\omega_i$.

The polarization is then $P^{(3)} = P^{(1)} + P^{(II)}$, where

$$P^{(I)} = S_1(r) \exp[-i(\omega_1 t + \omega_2 \tau)] \times \int_{-\infty}^{+\infty} dv \left( \int_{-\infty}^{+\infty} dt_1 \int_{0}^{+\infty} dt_2 \int_{0}^{+\infty} dt_3 \right) \times \exp[-i \theta_1(\mathbf{v})] \exp[-(\Gamma_{10} + i \Delta_1) t_3] \times \exp[-(\Gamma_{20} + i \Delta_2) t_2] \times \exp[-(\Gamma_{10} + i \Delta_1) t_1] \exp(-\alpha_2 |t_2 - \tau|),$$

$$P^{(II)} = S_2(r) \exp[-i(\omega_1 t + \omega_3 \tau)] \times \int_{-\infty}^{+\infty} dv \left( \int_{-\infty}^{+\infty} dt_1 \int_{0}^{+\infty} dt_2 \int_{0}^{+\infty} dt_3 \right) \times \exp[-i \theta_{II}(\mathbf{v})] \exp[-(\Gamma_{10} + i \Delta_1) t_3] \times \exp[-(\Gamma_{30} + i \Delta_3) t_2] \times \exp[-(\Gamma_{10} + i \Delta_1) t_1] \exp(-\alpha_3 |t_2 - \tau|),$$

where

$$S_1(r) = -\frac{i N \mu_1^2 \mu_2^2}{\hbar^3} e^{i \epsilon_1 \epsilon_2(\epsilon_2')^*} \times \exp[i(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_2') \cdot \mathbf{r}],$$

$$S_2(r) = -\frac{i N \mu_1^2 \mu_3^2}{\hbar^3} e^{i \epsilon_1 \epsilon_2(\epsilon_2')^*} \times \exp[i(\mathbf{k}_1 + \mathbf{k}_3 - \mathbf{k}_3') \cdot \mathbf{r}],$$

$$\theta_1(\mathbf{v}) = \mathbf{v} \cdot [\mathbf{k}_1(t_1 + t_2 + t_3) + \mathbf{k}_2(t_2 + t_3) - \mathbf{k}_3'(t_3)],$$

$$\theta_{II}(\mathbf{v}) = \mathbf{v} \cdot [\mathbf{k}_1(t_1 + t_2 + t_3) + \mathbf{k}_3(t_2 + t_3) - \mathbf{k}_3'(t_3)].$$

$\mu_1$, $\mu_2$, and $\mu_3$ are the dipole-moment matrix elements between $|0\rangle$ and $|1\rangle$, $|1\rangle$ and $|2\rangle$, and $|1\rangle$ and $|3\rangle$ and $\Gamma_{10}$, $\Gamma_{20}$, and $\Gamma_{30}$ are the transverse relaxation rates of the transitions from $|0\rangle$ to $|1\rangle$, $|0\rangle$ to $|2\rangle$, and $|0\rangle$ to $|3\rangle$, respectively; $\Delta_1 = \Omega_1 - \omega_1$, $\Delta_2 = \Omega_2 - \omega_2$, and $\Delta_3 = \Omega_3 - \omega_3$.

We now consider the case that beams 2 and 3 are narrow band, so $\alpha_2, \alpha_3 \ll \Gamma_{10}, \Gamma_{20}, \Gamma_{30}$. For simplicity, here we neglect the Doppler effect. Performing the tedious integration, we find that the beat density then becomes

$$I(\tau) \propto |P^{(3)}|^2 \propto \left[ B_1 \right]^2 - \frac{1}{(\Gamma_{10} + i \Delta_1)(\Gamma_{10} + \Gamma_{20} + i \Delta_2)} + \left[ B_2 \right]^2 - \frac{1}{(\Gamma_{10} + i \Delta_1)(\Gamma_{10} + \Gamma_{30} + i \Delta_3)} + \left[ B_1 \right]^2 \exp(-2 \alpha_2 |\tau|) + |\eta| \left[ B_2 \right]^2 \exp(-2 \alpha_3 |\tau|) + \exp[-(\alpha_2 + \alpha_3) |\tau|] \left( \eta B_1 B_2^* \right) + \exp[-i (\alpha_3 - \alpha_2) |\tau|] \eta \left( \eta B_1 B_2^* \right) \times \exp[i (\alpha_3 - \alpha_2) |\tau|],$$

with

$$B_1 = \frac{1}{(\Gamma_{10} + i \Delta_1 + i \Delta_2)(\Gamma_{10} + \Gamma_{20} + i \Delta_2)},$$

$$B_2 = \frac{1}{(\Gamma_{10} + i \Delta_1 + i \Delta_3)(\Gamma_{10} + \Gamma_{30} + i \Delta_3)},$$

$$\eta = \frac{S_2(r)}{S_1(r)} = \frac{\mu_3^2}{\mu_2^2} \left( \frac{\epsilon_3 \epsilon_3'^*}{\epsilon_2 \epsilon_2'^*} \right),$$

which is spatially dependent.

Relation (5) consist of three parts. The first and second terms, which depend on the fourth-order coherence function and originate from the amplitude fluctuation of the chaotic field, are independent of the relative time delay between beams 2 and 3. The third and fourth terms, which depend on the fourth-order coherence function and originate from the phase fluctuation of the chaotic field, indicate an exponential decay of the beat signal as $|\tau|$ increases. The fifth and sixth terms, which depend on the second-order coherence function and are determined by the laser line shape, give rise to the modulation of the beat signal.

Relation (5) indicates that beat signal oscillates not only temporally but also spatially, with a period $2 \pi / \left( |\mathbf{k}_2 - \mathbf{k}_3'| - |(\mathbf{k}_3 - \mathbf{k}_3')| \right)$ along the direction $|\mathbf{k}_2 - \mathbf{k}_3'| - |(\mathbf{k}_3 - \mathbf{k}_3')|$, which is almost perpendicular to the propagation direction of the beat signal. Physically, the polarization-beat model assumes that both of the pump beams are plane waves. Therefore two two-photon FWM signals, which propagate along $\mathbf{k}_1 = \mathbf{k}_2 - \mathbf{k}_3'$ and $\mathbf{k}_{32} = \mathbf{k}_3 - \mathbf{k}_3' + \mathbf{k}_1$, respectively, are plane waves also. Because the two two-photon FWM signals propagate along slightly different directions, the interference between them leads to spatial oscillation. However, the spatial dependence in $\eta$ can be neglected in a typical experiment. Therefore,

$$\eta = \frac{\mu_3^2}{\mu_2^2} \left( \frac{\epsilon_3 \epsilon_3'^*}{\epsilon_2 \epsilon_2'^*} \right).$$

Relation (5) only indicates that beat signal modulates with a frequency $\omega_3 - \omega_2$ as $\tau$ is varied. In this case $\omega_2$ and $\omega_3$ are tuned to the resonant frequencies of the transitions from $|1\rangle$ to $|2\rangle$ and from $|1\rangle$ to $|3\rangle$, respectively; then the modulation frequency equals $\Omega_3 - \Omega_2$. In the other words, we can obtain beating between the resonant frequencies of a four-level system. A Doppler-free precision can be achieved in the measurement of $\Omega_3 - \Omega_2$.5,6

3. PBFS IN A DOPPLER-BROADENED SYSTEM

The beat signal can be calculated from a different viewpoint. In the Doppler-broadened limit (i.e., $k_1 u \rightarrow \infty$), we have
\[
\int_{-\infty}^{\infty} dv w(v) \exp[-i \theta_1(v)] = \frac{2\sqrt{\pi}}{k_i u} \delta(t_1 + t_2 + t_3 - \xi_1 t_2), \tag{6}
\]
\[
\int_{-\infty}^{\infty} dv w(v) \exp[-i \theta_2(v)] = \frac{2\sqrt{\pi}}{k_i u} \delta(t_1 + t_2 + t_3 - \xi_2 t_2), \tag{7}
\]
where \(\xi_1 = k_2/k_1\) and \(\xi_2 = k_3/k_1\). We assume that \(\xi_1 > 1\) and \(\xi_2 > 1\). When we substitute relations (6) and (7) into relations (3) and (4) we obtain
\[
\langle |P^3|^2 \rangle = \langle |P^\Pi|^2 \rangle. \tag{8}
\]
We first consider the case that beams 2 and 3 are narrow band, so \(\alpha_2, \alpha_3 \ll \Gamma_20, \Gamma_30\). Performing a tedious integration yields the beat signal intensity
\[
I(\tau) \propto \langle |P^3|^2 \rangle
\]
\[
= \frac{(\xi_1 - 1)}{(\Gamma_20 - \Gamma_10 + i\Delta_2^a)^2} \times \left[ \frac{(\xi_1 - 1)^2 + 1}{2(\Gamma_20 - \Gamma_10)[(\xi_1 + 1)(\Gamma_20 - \Gamma_10) + i\Delta_2^a(\xi_1 - 1)]} \right]
\]
\[
+ \frac{\xi_1^2 - \xi_1 + 1}{(\Gamma_20 - \Gamma_10 + i\Delta_2^a)^2}
\]
\[
+ \frac{1}{2(\Gamma_20 - \Gamma_10)(\Gamma_20 - \Gamma_10 - i\Delta_2^a)}
\]
\[
- \frac{\xi_2 - 1}{2[(\Gamma_30 - \Gamma_10)^2 + (\Delta_3^a)^2]} + \frac{|\eta|^2(\xi_2 - 1)}{(\Gamma_30 - \Gamma_10 + i\Delta_3^a)^2}
\]
\[
\times \left[ \frac{(\xi_2 - 1)^2 + 1}{2(\Gamma_30 - \Gamma_10)[(\xi_2 + 1)(\Gamma_30 - \Gamma_10) + i\Delta_3^a(\xi_2 - 1)]} \right]
\]
\[
+ \frac{\xi_2^2 - \xi_2 + 1}{(\Gamma_30 - \Gamma_10 + i\Delta_3^a)^2}
\]
\[
+ \frac{1}{2(\Gamma_30 - \Gamma_10)(\Gamma_30 - \Gamma_10 - i\Delta_3^a)}
\]
\[
- \frac{\xi_3 - 1}{2[(\Gamma_30 - \Gamma_10)^2 + (\Delta_3^a)^2]} + \frac{|\eta|^2(\xi_3 - 1)}{(\Gamma_30 - \Gamma_10 + i\Delta_3^a)^2}
\]
\[
\times \left[ \frac{(\xi_3 - 1)^2}{[(\Gamma_30 - \Gamma_10)^2 + (\Delta_3^a)^2]} \exp(-2\alpha_2|\tau|) \right]
\]
\[
+ \frac{|\eta|^2(\xi_3 - 1)^2}{[(\Gamma_30 - \Gamma_10)^2 + (\Delta_3^a)^2]} \exp(-2\alpha_3|\tau|)
\]
\[
+ \exp(-\alpha_2 + \alpha_3)|\tau| q \exp(-i(\omega_3 - \omega_2)|\tau|)
\]
\[
+ q^* \exp[i(\omega_3 - \omega_2)|\tau|], \tag{9}
\]
where
\[
q = \frac{\eta(\xi_1 - 1)(\xi_2 - 1)}{[(\Gamma_30 - \Gamma_10 - i\Delta_3^a)^2(\Gamma_30 - \Gamma_10 + i\Delta_3^a)^2]}
\]
\[
\Gamma_20^a = \Gamma_20 + \xi_1\Gamma_10, \quad \Delta_2^a = \Delta_2 + \xi_1\Delta_1,
\]
\[
\Gamma_30^a = \Gamma_30 + \xi_2\Gamma_10, \quad \Delta_3^a = \Delta_3 + \xi_2\Delta_1.
\]
Relation (9) is consistent with relation (5).
We now consider the case that beams 2 and 3 are broadband, so \(\alpha_2, \alpha_3 \gg \Gamma_20, \Gamma_30\). In this case the beat signal rises to its maximum quickly and then decays with a time constant determined mainly by the transverse relaxation times of the system. Although the beat signal modulation is complicated in general, at the tail of the signal (i.e., \(\tau \gg \alpha_2^{-1}, \tau \gg \alpha_3^{-1}\)) we have
\[
(i) \tau > 0
\]
\[
I(\tau) \propto \langle |P^3|^2 \rangle
\]
\[
= \frac{(\xi_1 - 1)[\alpha_2^2 + (\Delta_2^a)^2 - 2i\alpha_2\Delta_2^a]}{(2\Gamma_20 - \Gamma_10)^2[\alpha_2^2 + (\Delta_2^a)^2]}
\]
\[
+ \frac{(\xi_2 - 1)[\alpha_3^2 + (\Delta_3^a)^2 - 2i\alpha_3\Delta_3^a]}{(2\Gamma_30 - \Gamma_10)^2[\alpha_3^2 + (\Delta_3^a)^2]}
\]
\[
+ \frac{2\Gamma_20 - \Gamma_10}{\alpha_2^2 + (\Delta_2^a)^2}
\]
\[
- \frac{4(\xi_1 - 1)\alpha_2^2\Delta_2^a\tau^2}{\alpha_2^2 + (\Delta_2^a)^2}
\]
\[
+ \frac{4(\xi_2 - 1)\alpha_3^2\Delta_3^a\tau^2}{\alpha_3^2 + (\Delta_3^a)^2}
\]
\[
- \frac{4(\xi_1 - 1)(\xi_2 - 1)\alpha_2\alpha_3\Delta_1^a\tau^2}{\alpha_2^2 + (\Delta_2^a)^2 \alpha_3^2 + (\Delta_3^a)^2}
\]
\[
\times \exp[-(\Gamma_20 - \Gamma_10)|\tau|]
\]
\[
\times \{ \eta \exp[-i(\Omega_3 - \Omega_2)\tau - i(\xi_2 - \xi_1)\Delta_1^a\tau]
\]
\[
+ \eta^* \exp[i(\Omega_3 - \Omega_2)\tau + i(\xi_2 - \xi_1)\Delta_1^a\tau] \} \tag{10}
\]
Relation (10) also consists of three parts. The first and second terms, which depend on the fourth-order coherence function and originate from the amplitude fluctuation of the chaotic field, are independent of the relative time delay \(\tau\). The third and fourth terms, which depend on the fourth-order coherence function and originate from the phase fluctuation of the chaotic field, indicate an exponential decay of the beat signal as \(|\tau|\) increases. The fifth and sixth terms, which depend on the second-order coherence function and are determined by the laser line shape, give rise to the modulation of the beat signal. Relation (10) indicates that the modulation frequency of the beat signal equals \(\Omega_3 - \Omega_2\) when \(\Delta_1 = 0\). The overall accuracy of using PBFS with broadband lights to measure the energy-level difference between two excited states is limited by the homogeneous linewidths.\(^5\,\text{6}\)
(ii) \( \tau < 0 \)

\[
I(\tau) \propto \langle |P^3| \rangle^2 = \left( \frac{\xi_1 - 1}{4(\Gamma_{20} - \Gamma_{10})^2} + \frac{|\eta|^2 \alpha_2^2 (\xi_2 - 1)}{4 \alpha_3^2 (\Gamma_{30} - \Gamma_{10})^2} \right) + \tau^3 (\xi_1 - 1)^2 \exp\{ -2(\Gamma_{20} - \Gamma_{10}) |\tau| \} + \tau^2 (\xi_1 - 1)^2 \exp\{ -2(\Gamma_{30} - \Gamma_{10}) |\tau| \} + \tau^3 (\xi_2 - 1)^2 \exp\{ -2(\Gamma_{30} + \Gamma_{20}) |\tau| \} \times \exp\{ -i(\Omega_3 - \Omega_2) \tau + i(\xi_2 - \xi_1) \Delta_1 \tau \}
\]

Relation (11) is consistent with relation (5). Therefore the requirement for the existence of a \( \tau \)-dependent beat signal for \( \tau < 0 \) is that the phase-correlated subpulses in beams 2 and 3 overlap temporally. Because beams 2 and 3 are mutually coherent, the temporal behavior of the beat signal should coincide with the case when beams 2 and 3 are nearly monochromatic.

4. PHOTON ECHO

It is interesting to understand the underlying physics in PBFS with incoherent light. Much attention has been paid recently to the study of various ultrafast phenomena by use of incoherent light sources.7,12 For the phase-matching condition \( k_3 - k_3' + k_1 \) and \( k_3 - k_3' + k_1 \) two sum-frequency trilevel echoes exist for perturbation chains (I) and (II).9 In the Doppler-broadened limit (i.e., \( k_1 u \rightarrow \infty \)), if we assume that beam 2 (beam 3) has a Gaussian line shape, we have

\[
\langle u_i(t_1) u_i^*(t_2) \rangle = \exp\left\{ -\frac{\alpha_i}{2\sqrt{\ln 2}} (t_1 - t_2)^2 \right\} = \exp\{ -[\beta_i (t_1 - t_2)]^2 \}, \quad i = 2, 3.
\]

We now consider the case that beams 2 and 3 are broadband, so \( \alpha_2, \alpha_3 \gg \Gamma_{20}, \Gamma_{30} \). Then

\[
\langle u_i(t_1) u_i^*(t_2) \rangle = \exp\{ -[\beta_i (t_1 - t_2)]^2 \] \approx \frac{\sqrt{\pi}}{\beta_i} \delta(t_1 - t_2), \quad i = 2, 3. \quad (12)
\]

When we substitute relations (11) and (12) into Eq. (8), we obtain the relations that follow.

(i) \( \tau > 0 \)

\[
I(\tau) \propto \langle |P^3| \rangle^2 = \left( \frac{\xi_1 - 1}{4(\Gamma_{20} - \Gamma_{10})^2} + \frac{|\eta|^2 \alpha_2^2 (\xi_2 - 1)}{4 \alpha_3^2 (\Gamma_{30} - \Gamma_{10})^2} \right) + \tau^2 (\xi_1 - 1)^2 \exp\{ -2(\Gamma_{20} - \Gamma_{10}) |\tau| \} + \tau^3 (\xi_1 - 1)^2 \exp\{ -2(\Gamma_{30} - \Gamma_{10}) |\tau| \} + \tau^3 (\xi_2 - 1)^2 \exp\{ -2(\Gamma_{30} + \Gamma_{20}) |\tau| \} \times \exp\{ -i(\Omega_3 - \Omega_2) \tau + i(\xi_2 - \xi_1) \Delta_1 \tau \}
\]

This relation is consistent with relation (10).

(ii) \( \tau < 0 \)

When \( \tau < 0 \), a photon echo does not exist for perturbation chains (I) and (II). The requirement for the existence of a \( \tau \)-dependent beat signal for \( \tau < 0 \) is that the phase-correlated subpulses in beams 2 and 3 overlap temporally. Because beams 2 and 3 are mutually coherent, the temporal behavior of the beat signal should coincide with that when beams 2 and 3 are nearly monochromatic. Therefore this case is consistent with relation (5).

We assumed in the above calculation that the laser sources are a chaotic field. A chaotic field, which is used to describe a multimode laser source, is characterized by the fluctuation of both the amplitude and the phase of the field. Another commonly used stochastic model is the phase-diffusion model, which is used to describe an amplitude-stabilized laser source. This model assumes that the amplitude of the laser field is a constant, while its phase fluctuates as random process. If the lasers have Lorentzian line shape, the fourth-order coherence function is

\[
\langle u_i(t_1) u_i(t_2) u_i^*(t_3) u_i^*(t_4) \rangle = \exp\{ -\alpha_i (|t_1 - t_3| + |t_2 - t_4| + |t_1 - t_4| + |t_2 - t_3| + |t_2 - t_4|) \} \times \exp\{ \alpha_i (|t_1 - t_2| + |t_3 - t_4|) \}, \quad i = 2, 3. \quad (14)
\]

We now consider the case that beams 2 and 3 are broadband, so \( \alpha_2, \alpha_3 \gg \Gamma_{20}, \Gamma_{30} \). Then

\[
\langle u_i(t_1) u_i^*(t_2) \rangle = \exp\{ -\alpha_i (|t_1 - t_2|) \] \approx \frac{2}{\alpha_i} \delta(t_1 - t_2), \quad i = 2, 3. \quad (15)
\]

When we substitute Eqs. (14) and (15) into Eq. (8), we obtain the relations that follow.
(i) $\tau > 0$

\[
I(\tau) \propto \langle |P^{3/2}|^2 \rangle = \tau^2 (\xi_1 - 1)^2 \exp[-2(\Gamma_2 \alpha - \Gamma_10)|\tau|] \\
+ |\eta|^2 \tau^2 (\xi_2 - 1)^2 \frac{\alpha_2}{\alpha_3} \exp[-2(\Gamma_3 \alpha - \Gamma_10)|\tau|] \\
+ \tau^2 (\xi_1 - 1)(\xi_2 - 1) \frac{\alpha_2}{\alpha_3} \exp[-(\Gamma_2 + \Gamma_2 \alpha - 2\Gamma_10)|\tau|] \\
\times \{ \eta \exp[-i(\Omega_3 - \Omega_2)\tau - i(\xi_2 - \xi_1)\Delta_1 \tau] \\
+ \eta^* \exp[i(\Omega_3 - \Omega_2)\tau + i(\xi_2 - \xi_1)\Delta_1 \tau] \}. \quad (16)
\]

(ii) $\tau < 0$

When $\tau < 0$, a photon echo does not exist for perturbation chains (I) and (II). Relation (16) consist of two parts. The first and second terms, which depend on the fourth-order coherence function and originate from the phase fluctuation of the phase-diffusion field, indicate an exponential decay of the beat signal as $|\tau|$ increases. The third and fourth terms, which depend on the second-order coherence function and are determined by the laser line shape, give rise to the modulation of the beat signal. This case is consistent with the result of the second-order coherence-function theory; the constant-background contribution was ignored in the analysis of Ref. 5. Therefore the fourth-order coherence function theory of chaotic field is of vital importance in PBFS.

5. EXPERIMENT AND RESULT

We performed PBFS in sodium vapor, where the ground state $3S_{1/2}$, the intermediate state $3P_{1/2}$, and two excited states $6S_{1/2}$ and $5D_{3/2}$ form a four-level system. Three dye lasers (DL1, DL2, and DL3) pumped by the second harmonic of a Quanta-Ray YAG laser were used to generate frequencies at $\omega_1$, $\omega_2$, and $\omega_3$, respectively. DL1–DL3 had linewidths of 0.01 nm and pulse widths of 5 ns. DL1 was tuned to 589.6 nm, the wavelength of the $3S_{1/2}$–$3P_{1/2}$ transition, DL2 was tuned to 514.9 nm, the wavelength of the $3P_{1/2}$–$6S_{1/2}$ transition, and DL3 was tuned to 497.9 nm, the wavelength of the $3P_{1/2}$–$5D_{3/2}$ transition. A beam splitter was used to combine the $\omega_2$ and $\omega_3$ components derived from DL2 and DL3, respectively, for beams 2 and 3, which intersected in the oven containing the sodium vapor. Time delay $\tau$ between beams 2 and 3 could be varied. Beam 1, which propagated along the direction opposite that of beam 2, was derived from DL1. All the incident beams were linearly polarized in the same direction. The beat signal had the same polarization as the incident beams, propagated along a direction almost opposite that of beam 3. It was detected by a photodiode.

We first performed a degenerate FWM experiment with beams 1, 2, and 3 consisting of the $\omega_1$ frequency component. From the degenerate FWM spectrum we tuned $\omega_1$ to the resonant frequency $\Omega_1$. We then performed the first nondegenerate FWM experiment with beams 2 and 3 consisting of only the $\omega_2$ frequency component. Figure 3 shows the relation between the signal intensity and the relative time delay. We performed a second nondegenerate FWM experiment in which beams 2 and 3 consisted of only the $\omega_3$ frequency component. Figure 4 shows the relation between the signal intensity and the relative time delay. After that, we performed a PBFS experiment by measuring the beat signal intensity as a function of the relative time delay when beams 2 and 3 consist of both frequencies $\omega_2$ and $\omega_3$. Figure 5 shows the result of the beat experiment in which $\tau$ is varied for a range of 5 ps. The beat signal intensity modulates sinusoidally with period 50.27 fs. One can obtain modulation frequency more directly by performing a Fourier transformation of the PBFS data. Figure 6 presents the Fourier spectrum of the data in which $\tau$ is varied for a range of 5 ps. Then we obtain the modulation frequency $1.25 \times 10^{14}$ s$^{-1}$, corre-
sponding to beat between the resonant frequencies of the transitions from $3P_{1/2}$ to $6S_{1/2}$ and from $3P_{3/2}$ to $5D_{3/2}$.

Now we discuss the difference between PBFS and DeBeer’s UMS with self-diffraction geometry from a physical viewpoint. The frequencies and the wave vectors of DeBeer’s UMS signal are $\omega_{s1} = 2 \omega_1 - \omega_2$ and $\omega_{s2} = 2 \omega_2 - \omega_1$ and $k_{s1} = 2k_1' - k_1$ and $k_{s2} = 2k_2' - k_2$, respectively, which means that a photon is absorbed from each of the two mutually pumped beams. On the other hand, the frequencies and the wave vectors of the PBFS signal are $\omega_{s1} = \omega_2 - \omega_1 + \omega_3$ and $\omega_{s2} = \omega_3 - \omega_1$ and $k_{s1} = k_2 - k_2' + k_1$ and $k_{s2} = k_3 - k_3' + k_1$, respectively; therefore photons are absorbed from and emitted to the mutually correlated beams 2 and 3, respectively. This difference between the PBFS and DeBeer’s UMS has a profound influence on the field-correlation effects. We note that the roles of the two pump beams are interchangeable in DeBeer’s UMS. This interchangeable feature also makes the second-order coherence function theory fail in DeBeer’s UMS. In light of the fact that $\langle u(t_1)u(t_2) \rangle = 0$, the absolute square of the stochastic average of the polarization $|\langle P_{(3)} \rangle|^2$ cannot be used to describe the temporal behavior of DeBeer’s UMS. Our fourth-order theory is of vital importance in DeBeer’s UMS.

In conclusion, we have employed chaotic and phase-diffusion models to study the effect of laser-fourth-order coherence on PBFS in a four-level system. We found that the temporal behavior of the beat signal depends on the stochastic properties of the lasers and on the transverse relaxation rate of the atomic energy-level system. The different roles of phase fluctuation and amplitude fluctuation have been pointed out. Cases in which the pump beams have either a narrow-band or a broadband line-width were considered, and it was found that for both cases a Doppler-free precision in the measurement of the energy level-difference between two states that are dipolar forbidden from the ground state can be achieved. It is worth mentioning that the asymmetric behavior of the polarization beat signal owing to the dispersion of the optical components in a four-level system does not affect the overall accuracy of using PBFS to measure the energy-level difference. Furthermore, PBFS can tolerate small perturbations of the optical path caused by mechanical vibration, dispersion, and distortion of the optical components as long as these perturbations are small compared with $c/|\omega_3 - \omega_2|$.

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Y. Zhang’s e-mail address is yp.zhang@263.net.

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