PHYSICAL REVIEW A, VOLUME 61, 053819

Effects of field correlation on polarization beats

Yanpeng Zhang,* Liqun Sun, Tiantong Tang, and Panming Fu
Department of Electronic Science and Technology, Xi'an Jiaotong University, Xi'an 710049, China
(Received 13 January 1999; revised manuscript received 21 September 1999; published 18 April 2000)

We have employed a second-order coherence function theory to study the effect of laser coherence on polarization beats in four-level system (PBFS). It is found that the temporal behavior of the beat signal depends on the stochastic properties of the lasers and transverse relaxation rate of the transition. The cases that pump beams have either narrow band or broadband linewidth are considered and it has been found that for both cases a Doppler-free precision in the measurement of the energy-level splitting between two excited states which are dipolar forbidden from the ground state can be achieved. We have also studied the asymmetric behavior of the polarization beats, and attributed this asymmetry to the shift of the zero time delay which is due to the dispersion of the optical components.

PACS number(s): 42.50.Md, 42.65.Hw, 32.90.+a

I. INTRODUCTION

Polarization beats, which originate from the interference between the macroscopic polarizations, have attracted a lot of attention recently. It is closely related to the quantum beat spectroscopy, which appears in the conventional time-resolved fluorescence and in the time-resolved nonlinear laser spectroscopy. Up to now, quantum beat spectroscopy is applied to the quasi-two-level [1,2] and cascade three-level systems [3]. In the quasi-two-level, the excited and the ground states consist of sublevel structures. A quantum beat manifests itself as an oscillation of the signal with frequency corresponding to the energy-level splittings. For example, Debeer *et al.* [1] performed the first ultrafast modulation spectroscopy (UMS) experiment in sodium vapor. The beating signal exhibits 1.9 ps modulation corresponding to the sodium *D*-line splitting when the time delay between two double-frequency pump beams increases. Fu *et al.* [2] then analyzed the UMS with phase-conjugate geometry in a Doppler-broadened system. They found that a Doppler-free precision in the measurement of the energy-level splitting could be achieved. Based on the interference between one-photon and two-photon processes, the UMS technique has also been applied to a cascade three-level system [3]. Ultrafast modulation spectroscopy in cascade three-level shows beating between the resonant frequencies of a cascade three-level system. If the energy separation between the ground and the intermediate states is well known, then from the beating the energy separation between the intermediate and excited states can be deduced. Based on the interference between two two-photon processes, we extend the UMS technique to the four-level system.

In this paper, we report the effects of field-correlation on polarization beat spectroscopy in a four-level system. We assumed 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, while its phase fluctuates as a random process. We present a second-order coherence function theory to elucidate the physics of the basic features of the effects of field correlation on polarization beats in a four-level system. We also studied the asymmetric behavior of the polarization beats [4–6], and attribute this asymmetry to the shift of the zero time delay which is due to the dispersion of the optical components. If PBFS is employed for the energy-level difference measurement, 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 have also investigated the relationship between PBFS and other Doppler-free techniques in frequency and in time domains. It is found that PBFS is closely related to the two-photon absorption spectroscopy with a resonant intermediate state [3] and the sum-frequency trilevel photon echo [7] when the pump beams are narrow band and broadband, respectively. However, it possesses the main advantages of these techniques in the frequency domain and in the time domain.

II. BASIC THEORY

PBFS is a polarization beat phenomenon originating from the interference between two two-photon processes. Let us consider a four-level system (Fig. 1) with a ground state $|0\rangle$, an intermediate state $|1\rangle$ and two excited states $|2\rangle$ and $|3\rangle$. States between $|0\rangle$ and $|1\rangle$ and between $|1\rangle$ and $|2\rangle$ ($|3\rangle$) are coupled by dipolar transition with resonant frequencies Ω_1

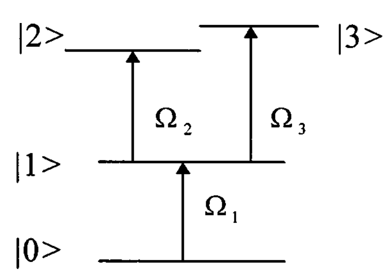

FIG. 1. Four-level configuration to be treated in PBFS.

*FAX: +86-29-3237910. Electronic address: yp.zhang@263.net
Author to whom correspondence should be addressed.

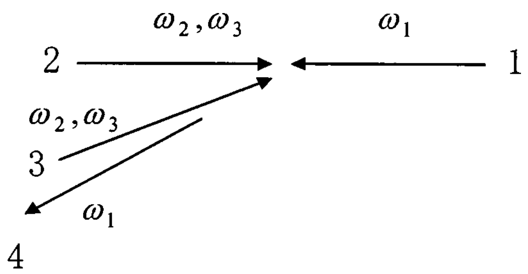

FIG. 2. Schematic diagram of the geometry of PBFS.

and $\Omega_2(\Omega_3)$, respectively, while states between $|2\rangle$ and $|3\rangle$ and between $|0\rangle$ and $|2\rangle(|3\rangle)$ are dipolar forbidden. We consider in this four-level system a double-frequency time-delay four-wave mixing (FWM) experiment in which beams 2 and 3 consist of two frequency components ω_2 and ω_3, while beam 1 has frequency ω_1 (Fig. 2). We assume that $\omega_1 \approx \Omega_1$ and $\omega_2 \approx \Omega_2(\omega_3 \approx \Omega_3)$, therefore ω_1 and $\omega_2(\omega_3)$ will drive the transitions from $|0\rangle$ to $|1\rangle$ and from $|1\rangle$ to $|2\rangle(|3\rangle)$, respectively. In this double-frequency time-delay FWM, beam 1 with frequency ω_1 and the $\omega_2(\omega_3)$ frequency component of beam 2 induce coherence between $|0\rangle$ and $|2\rangle(|3\rangle)$ by two-photon transition, which is then probed by the $\omega_2(\omega_3)$ frequency component of beam 3. These are two-photon FWM, with a resonant intermediate state and the frequency of the signal equals ω_1. We are interested in the dependence of the beating signal intensity on the relative time delay between 2 and 3.

The complex electric fields of beam 2, E_{p2}, and beam 3, E_{p3}, can be written as

$$E_{P_2} = \varepsilon_2 u_2(t) \exp[i(\vec{k}_2 \cdot \vec{r} - \omega_2 t)] + \varepsilon_3 u_3(t) \times \exp[i(\vec{k}_3 \cdot \vec{r} - \omega_3 t)],$$

$$E_{P_3} = \varepsilon_2' u_2(t-\tau) \exp[i(\vec{k}_2' \cdot \vec{r} - \omega_2 t + \omega_2 \tau)] + \varepsilon_3' u_3(t-\tau+\delta\tau) \exp[i(\vec{k}_3' \cdot \vec{r} - \omega_3 t + \omega_3 \tau - \omega_3 \delta\tau)].$$

Here, $\varepsilon_i, \vec{k}_i$ ($\varepsilon_i', \vec{k}_i'$) are the constant field amplitude and the wave vector of the ω_i component in beam 2 (beam 3). $u_i(t)$ is a dimensionless statistical factor that contains phase and amplitude fluctuations. We assume that the $\omega_2(\omega_3)$ component of E_{p2} and E_{p3} comes from a single laser source and τ is the time delay of beam 3 with respect to beam 2. $\delta\tau$ denotes the difference in the zero time delay. On the other hand, the beam 1 is assumed to be a quasimonochromatic light, the complex electric fields of beam 1 can be written as $E_{P_1} = \varepsilon_1 \exp[i(\vec{k}_1 \cdot \vec{r} - \omega_1 t)]$.

We employ perturbation theory to calculate the density matrix elements. In the following perturbation chains: (I) $\rho_{00}^{(0)} \xrightarrow{\omega_1} \rho_{10}^{(1)} \xrightarrow{\omega_2} \rho_{20}^{(2)} \xrightarrow{-\omega_2} \rho_{10}^{(3)}$, (II) $\rho_{00}^{(0)} \xrightarrow{\omega_1} \rho_{10}^{(1)} \xrightarrow{\omega_3} \rho_{30}^{(2)} \xrightarrow{-\omega_3} \rho_{10}^{(3)}$. We obtain the third-order off-diagonal density matrix element $\rho_{10}^{(3)}$ which has wave vector $\vec{k}_2 - \vec{k}_2' + \vec{k}_1$ or $\vec{k}_3 - \vec{k}_3' + \vec{k}_1$, $\rho_{10}^{(3)} = \rho^{(\mathrm{I})} + \rho^{(\mathrm{II})}$. Here $\rho^{(\mathrm{I})}$ and $\rho^{(\mathrm{II})}$ corresponding to $\rho_{10}^{(3)}$ of the perturbation chain (I) and (II), respectively, are

$$\rho_{10}^{(\mathrm{I})} = \frac{-i\mu_1\mu_2^2}{\hbar^3} \varepsilon_1 \varepsilon_2 (\varepsilon_2')^* \exp\{i[(\vec{k}_1 + \vec{k}_2 - \vec{k}_2') \cdot \vec{r} - \omega_1 t - \omega_2 \tau]\} \int_0^\infty dt_3 \int_0^\infty dt_2 \int_0^\infty dt_1 \times \exp\{-i\vec{v} \cdot [\vec{k}_1(t_1+t_2+t_3) + \vec{k}_2(t_2+t_3) - \vec{k}_2' t_3]\} \times \exp[-(\Gamma_{10} + i\Delta_1)t_3] \exp[-(\Gamma_{20} + i\Delta_1 + i\Delta_2)t_2] \times \exp[-(\Gamma_{10} + i\Delta_1)t_1] u_2(t-t_2-t_3) u_2^*(t-t_3-\tau), \tag{1}$$

$$\rho_{10}^{(\mathrm{II})} = \frac{-i\mu_1\mu_3^2}{\hbar^3} \varepsilon_1 \varepsilon_3 (\varepsilon_3')^* \exp\{i[(\vec{k}_1 + \vec{k}_3 - \vec{k}_3') \cdot \vec{r} - \omega_1 t - \omega_3 \tau + \omega_3 \delta\tau]\} \int_0^\infty dt_3 \int_0^\infty dt_2 \int_0^\infty dt_1 \times \exp\{-i\vec{v} \cdot [\vec{k}_1(t_1+t_2+t_3) + \vec{k}_3(t_2+t_3) - \vec{k}_3' t_3]\} \times \exp[-(\Gamma_{10} + i\Delta_1)t_3] \exp[-(\Gamma_{30} + i\Delta_1 + i\Delta_3)t_2] \times \exp[-(\Gamma_{10} + i\Delta_1)t_1] u_3(t-t_2-t_3) \times u_3^*(t-t_3-\tau+\delta\tau). \tag{2}$$

Here, $\vec{v}$ is the atomic velocity; μ_1, μ_2, μ_3 is the dipole-moment matrix element between $|0\rangle$ and $|1\rangle$, $|1\rangle$ and $|2\rangle$, $|1\rangle$ and $|3\rangle$, $\Gamma_{10}, \Gamma_{20}, \Gamma_{30}$ is the transverse relaxation rate of the transition from $|0\rangle$ and $|1\rangle$, $|0\rangle$ to $|2\rangle$, $|0\rangle$ to $|3\rangle$; $\Delta_1 = \Omega_1 - \omega_1$, $\Delta_2 = \Omega_2 - \omega_2$, $\Delta_3 = \Omega_3 - \omega_3$.

The nonlinear polarization $P^{(3)}$ responsible for the phase-conjugate FWM signal is given by averaging over the velocity distribution function $W(\vec{v})$. Thus $P^{(3)} = N\mu_1 \int_{-\infty}^{+\infty} d\vec{v}\, w(\vec{v}) \rho_{10}^{(3)}(\vec{v})$, here, N is the density of atoms. For a Doppler-broadened atomic system, we have $w(\vec{v}) = [1/(u\sqrt{\pi})] \exp[-(\vec{v}/u)^2]$, here, $u = \sqrt{2k_B T/m}$ with m the mass of an atom, k_B Boltzmann's constant, and T the absolute temperature. The FWM signal is proportional to the average of the absolute square of $P^{(3)}$ over the random variable of the stochastic process $\langle |P^{(3)}|^2 \rangle$, which involves fourth-order coherence function of $u_i(t)$ in phase-conjugation geometry. While the FWM signal intensity in Debeer's self-diffraction geometry is related to the sixth-order coherence function of the incident fields. We assume that beam 2 (beam 3) is multimode thermal source. $u_i(t)$ has Gaussian statistics with its fourth-order coherence function satisfying [8]

$$\langle u_i(t_1) u_i(t_2) u_i^*(t_3) u_i^*(t_4) \rangle = \langle u_i(t_1) u_i^*(t_3) \rangle \langle u_i(t_2) u_i^*(t_4) \rangle + \langle u_i(t_1) u_i^*(t_4) \rangle \langle u_i(t_2) u_i^*(t_3) \rangle, \quad i = 2,3.$$

In the case where we are only interested in the τ-dependent part of the signal, the FWM signal intensity can be well approximated by the absolute square of the stochastic average of the polarization $|\langle P^{(3)}\rangle|^2$, which involves a second-order function of $u_i(t)$. Here we develop a second-order function theory to study the effect of laser coherence. This theory is valid when we are only interested in the τ-dependent part of the beat signal. Furthermore assuming that beam 2 (beam 3) has Lorentzian line shape, then we have

$$\langle u_i(t_1)u_i^*(t_2)\rangle = \exp(-\alpha_i|t_1-t_2|) \quad (i=2,3),$$

here, $\alpha_i=(1/2)\delta\omega_i$ with $\delta\omega_i$ the linewidth of the laser with frequency ω_i.

Then the stochastic average of the polarization is $\langle P^{(3)}\rangle = P^{(\mathrm{I})}+P^{(\mathrm{II})}$, where

$$\begin{aligned} P^{\mathrm{I}} = {} & S_1(\vec{r})\exp[-i(\omega_1 t+\omega_2\tau)] \\ & \times \int_{-\infty}^{+\infty} d\vec{v}\, w(\vec{v}) \int_0^\infty dt_3 \int_0^\infty dt_2 \int_0^\infty dt_1 \\ & \times \exp[-i\theta_{\mathrm{I}}(\vec{v})]\exp[-(\Gamma_{10}+i\Delta_1)t_3] \\ & \times \exp[-(\Gamma_{20}+i\Delta_2+i\Delta_1)t_2]\exp[-(\Gamma_{10}+i\Delta_1)t_1] \\ & \times \exp(-\alpha_2|t_2-\tau|), \end{aligned} \tag{3}$$

$$\begin{aligned} P^{(\mathrm{II})} = {} & S_2(\vec{r})\exp[-i(\omega_1 t+\omega_3\tau-\omega_3\delta\tau)] \\ & \times \int_{-\infty}^{+\infty} d\vec{v}\, w(\vec{v}) \int_0^\infty dt_3 \int_0^\infty dt_2 \int_0^\infty dt_1 \\ & \times \exp[-i\theta_{\mathrm{II}}(\vec{v})]\exp[-(\Gamma_{10}+i\Delta_1)t_3] \\ & \times \exp[-(\Gamma_{30}+i\Delta_3+i\Delta_1)t_2]\exp[-(\Gamma_{10}+i\Delta_1)t_1] \\ & \times \exp(-\alpha_3|t_2-\tau+\delta\tau|). \end{aligned} \tag{4}$$

Here,

$$S_1(\vec{r}) = -\frac{iN\mu_1^2\mu_2^2}{\hbar^3}\varepsilon_1\varepsilon_2(\varepsilon_2')^* \exp[i(\vec{k}_1+\vec{k}_2-\vec{k}_2')\cdot\vec{r}],$$

$$S_2(\vec{r}) = -\frac{iN\mu_1^2\mu_3^2}{\hbar^3}\varepsilon_1\varepsilon_2(\varepsilon_3')^* \exp[i(\vec{k}_1+\vec{k}_3-\vec{k}_3')\cdot\vec{r}],$$

$$\theta_{\mathrm{I}}(\vec{v}) = \vec{v}\cdot[\vec{k}_1(t_1+t_2+t_3)+\vec{k}_2(t_2+t_3)-\vec{k}_2' t_3],$$

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

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

$$\begin{aligned} I(\tau) \propto {} & |B_1|^2\exp(-2\alpha_2|\tau|) + |\eta B_2|^2\exp(-2\alpha_3|\tau-\delta\tau|) \\ & + \exp(-\alpha_2|\tau|)\exp(-\alpha_3|\tau-\delta\tau|)\{\eta B_1^* B_2 \\ & \times \exp[-i(\omega_3-\omega_2)\tau+i\omega_3\delta\tau] + \eta^* B_1 B_2^* \\ & \times \exp[i(\omega_3-\omega_2)\tau-i\omega_3\delta\tau]\}. \end{aligned} \tag{5}$$

with

$$B_1 = \frac{1}{(\Gamma_{10}+i\Delta_1)^2}\,\frac{1}{\Gamma_{20}+i(\Delta_2+\Delta_1)},$$

$$B_2 = \frac{1}{(\Gamma_{10}+i\Delta_1)^2}\,\frac{1}{\Gamma_{30}+i(\Delta_3+\Delta_1)},$$

and

$$\eta = \frac{S_2(\vec{r})}{S_1(\vec{r})} \approx \frac{\mu_3^2}{\mu_2^2}\left[\frac{\varepsilon_3(\varepsilon_3')^*}{\varepsilon_2(\varepsilon_2')^*}\right].$$

Equation (5) indicates that beat signal modulates with a frequency $\omega_3-\omega_2$ as τ is varied. In this case that ω_2 and ω_3 are tuned to the resonant frequencies of the transitions from $|1\rangle$ to $|2\rangle$ and from $|1\rangle$ and $|3\rangle$, respectively, then the modulation frequency equals $\Omega_3-\Omega_2$. In 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 $\Omega_3-\Omega_2$ [2]. On the other hand, the temporal behavior of the beat signal is asymmetric with the maximum of the signal shifted from $\tau=0$. We attribute this asymmetry to the shift of the zero time delay which is due to the dispersion of the optical components.

III. PBFS IN A DOPPLER-BROADENED SYSTEM

The beat signal can be calculated from a different viewpoint. Under the Doppler-broadened limit (i.e., $k_1u\to\infty$), we have

$$\int_{-\infty}^{+\infty} d\vec{v}\, w(\vec{v})\exp[-i\theta_I(\vec{v})] \approx \frac{2\sqrt{\pi}}{k_1 u}\,\delta(t_1+t_2+t_3-\xi_1 t_2), \tag{6}$$

$$\int_{-\infty}^{+\infty} d\vec{v}\, w(\vec{v})\exp[-i\theta_{II}(\vec{v})] \approx \frac{2\sqrt{\pi}}{k_1 u}\,\delta(t_1+t_2+t_3-\xi_2 t_2), \tag{7}$$

here, $\xi_1=k_2/k_1$, $\xi_2=k_3/k_1$. We assume $\xi_1>1$, $\xi_2>1$, $\delta\tau>0$. When we substitute Eqs. (6), (7) into Eqs. (3), (4) we obtain (i) $\tau>\delta\tau$,

$$\langle P^{(3)}\rangle = P^{(\mathrm{I})}+P^{(\mathrm{II})}=\left(\frac{2\sqrt{\pi}}{k_1 u}\right)S_1(\vec{r})\exp[-i(\omega_1 t+\omega_2\tau)](\xi_1-1)\left\{\frac{\exp(-\alpha_2|\tau|)}{(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+i\Delta_2^a)^2}+\exp[-(\Gamma_{20}^a-\Gamma_{10}+i\Delta_2^a)|\tau|]\right.$$
$$\times\left[\frac{-\tau(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+i\Delta_2^a)-1}{(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+i\Delta_2^a)^2}+\frac{\tau(\Gamma_{20}^a-\Gamma_{10}+\alpha_2+i\Delta_2^a)+1}{(\Gamma_{20}^a-\Gamma_{10}+\alpha_2+i\Delta_2^a)^2}\right]\Bigg\}+\frac{2\sqrt{\pi}}{k_1 u}S_2(\vec{r})\exp[-i(\omega_1 t+\omega_3\tau-\omega_3\delta\tau)]$$
$$\times(\xi_2-1)\left\{\frac{\exp(-\alpha_3|\tau-\delta\tau|)}{(\Gamma_{30}^a-\Gamma_{10}-\alpha_3+i\Delta_3^a)^2}+\exp[-(\Gamma_{30}^a-\Gamma_{10}+i\Delta_3^a)|\tau-\delta\tau|]\left[\frac{-(\tau-\delta\tau)(\Gamma_{30}^a-\Gamma_{10}-\alpha_3+i\Delta_3^a)-1}{(\Gamma_{30}^a-\Gamma_{10}-\alpha_3+i\Delta_3^a)^2}\right.\right.$$
$$\left.\left.+\frac{(\tau-\delta\tau)(\Gamma_{30}^a-\Gamma_{10}+\alpha_3+i\Delta_3^a)+1}{(\Gamma_{30}^a-\Gamma_{10}+\alpha_3+i\Delta_3^a)^2}\right]\right\}. \tag{8}$$

Here, $\Gamma_{20}^\alpha=\Gamma_{20}+\xi_1\Gamma_{10}$, $\Delta_2^\alpha=\Delta_2+\xi_1\Delta_1$, $\Gamma_{30}^a=\Gamma_{30}+\xi_2\Gamma_{10}$, $\Delta_3^\alpha=\Delta_3+\xi_2\Delta_1$. (ii) $0<\tau<\delta\tau$,

$$\langle P^{(3)}\rangle = P^{(\mathrm{I})}+P^{(\mathrm{II})}=\left(\frac{2\sqrt{\pi}}{k_1 u}\right)S_1(\vec{r})\exp[-i(\omega_1 t+\omega_2\tau)]$$
$$\times(\xi_1-1)\left\{\frac{\exp(-\alpha_2|\tau|)}{(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+i\Delta_2^a)^2}\right.$$
$$+\exp[-(\Gamma_{20}^a-\Gamma_{10}+i\Delta_2^a)|\tau|]$$
$$\times\left[\frac{-\tau(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+i\Delta_2^a)-1}{(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+i\Delta_2^a)^2}\right.$$
$$\left.\left.+\frac{\tau(\Gamma_{20}^a-\Gamma_{10}+\alpha_2+i\Delta_2^a)+1}{(\Gamma_{20}^a-\Gamma_{10}+\alpha_2+i\Delta_2^a)^2}\right]\right\}$$
$$+\frac{2\sqrt{\pi}}{k_1 u}S_2(\vec{r})\exp[-i(\omega_1 t+\omega_3\tau-\omega_3\delta\tau)](\xi_2-1)$$
$$\times\exp(-\alpha_3|\tau-\delta\tau|)\frac{1}{(\Gamma_{30}^a-\Gamma_{10}-\alpha_3+i\Delta_3^a)^2}. \tag{9}$$

(iii) $\tau<0$

$$\langle P^{(3)}\rangle = P^{(\mathrm{I})}+P^{(\mathrm{II})}=\left(\frac{2\sqrt{\pi}}{k_1 u}\right)S_1(\vec{r})\exp[-i(\omega_1 t+\omega_2\tau)]$$
$$\times(\xi_1-1)\exp(-\alpha_2|\tau|)\frac{1}{(\Gamma_{20}^a-\Gamma_{10}-\alpha_2+i\Delta_2^a)^2}$$
$$+\frac{2\sqrt{\pi}}{k_1 u}S_2(\vec{r})\exp[-i(\omega_1 t+\omega_3\tau-\omega_3\delta\tau)]$$
$$\times(\xi_2-1)\exp(-\alpha_3|\tau-\delta\tau|)$$
$$\times\frac{1}{(\Gamma_{30}^a-\Gamma_{10}-\alpha_3+i\Delta_3^a)^2}. \tag{10}$$

We first consider the case that beams 2 and 3 are narrow band so that $\alpha_2\ll\Gamma_{20}$ and $\alpha_3\ll\Gamma_{30}$. The beat signal intensity is

$$I(\tau)\propto\frac{(\xi_1-1)^2\exp(-2\alpha_2|\tau|)}{[(\Gamma_{20}^\alpha-\Gamma_{10})^2+(\Delta_2^\alpha)^2]^2}$$
$$+\frac{|\eta|^2(\xi_2-1)^2\exp(-2\alpha_3|\tau-\delta\tau|)}{[(\Gamma_{30}^\alpha-\Gamma_{10})^2+(\Delta_3^\alpha)^2]^2}+\exp(-\alpha_2|\tau|)$$
$$\times\exp(-\alpha_3|\tau-\delta\tau|)\{q\exp[-i(\omega_3-\omega_2)\tau+i\omega_3\delta\tau]$$
$$+q^*\exp[i(\omega_3-\omega_2)\tau-i\omega_3\delta\tau]\}, \tag{11}$$

where

$$q=\frac{\eta(\xi_1-1)(\xi_2-1)}{[(\Gamma_{20}^a-\Gamma_{10})-i\Delta_2^a]^2[(\Gamma_{30}^a-\Gamma_{10})+i\Delta_3^a]^2}.$$

Equation (11) is consistent with Eq. (5).

We now consider the case that beams 2 and 3 are broadband so that $\alpha_2\gg\Gamma_{20}$ and $\alpha_3\gg\Gamma_{30}$. In this case, the beat signal rises to its maximum quickly and then decays with time constant mainly determined by the transverse relaxation times of the system. Although the beat signal modulation is complicated in general, we have from Eqs. (8), (9) that at the tail of the signal (i.e., $\tau\gg\alpha_2^{-1},\tau\gg\alpha_3^{-1}$) (i) $\tau>\delta\tau$,

$$I(\tau)\propto\left[\frac{\alpha_2(\xi_1-1)\tau}{\alpha_2^2+(\Delta_2^a)^2}\right]^2\exp[-2(\Gamma_{20}^a-\Gamma_{10})|\tau|]$$
$$+|\eta|^2\left[\frac{\alpha_3(\xi_2-1)(\tau-\delta\tau)}{\alpha_3^2+(\Delta_3^a)^2}\right]^2$$
$$\times\exp[-2(\Gamma_{30}^a-\Gamma_{10})|\tau-\delta\tau|]+\left[\frac{\alpha_2(\xi_1-1)\tau}{\alpha_2^2+(\Delta_2^a)^2}\right]$$
$$\times\left[\frac{\alpha_3(\xi_2-1)\tau}{\alpha_3^2+(\Delta_3^a)^2}\right]\tau(\tau-\delta\tau)\exp[-(\Gamma_{20}^a-\Gamma_{10})|\tau|]$$
$$\times\exp[-(\Gamma_{30}^a-\Gamma_{10})|\tau-\delta\tau|]\{\eta\exp[-i(\Omega_3-\Omega_2)\tau$$
$$-i(\xi_2-\xi_1)\Delta_1\tau+i(\Omega_3+\xi_2\Delta_1)\delta\tau]+\eta^*$$
$$\times\exp[i(\Omega_3-\Omega_2)\tau+i(\xi_2-\xi_1)\Delta_1\tau$$
$$-i(\Omega_3+\xi_2\Delta_1)\delta\tau]\}. \tag{12}$$

Equation (12) indicates that the modulation frequency of the beat signal equals $\Omega_3-\Omega_2$ when $\Delta_1=0$ and $\delta\tau=0$. The overall accuracy of using PBFS with broadband lights to measure the energy-level splitting between two excited states which are dipolar forbidden from the ground state is limited by the homogeneous linewidths [3]. On the other hand, the temporal behavior of the beat signal is asymmetric with the maximum of the signal shifted from $\tau=0$. We attribute this asymmetry to the shift of the zero time delay between beams 2 and 3 for the ω_2 and ω_3 frequency components which is due to the dispersion of the optical components. (ii) $0<\tau<\delta\tau$,

$$I(\tau)\propto\left[\frac{\alpha_2(\xi_1-1)\tau}{\alpha_2^2+(\Delta_2^a)^2}\right]^2\exp[-2(\Gamma_{20}^a-\Gamma_{10})|\tau|]. \tag{13}$$

Equation (13) indicates that the beat signal becomes the FWM signal intensity which beams 2 and 3 only consist of ω_2 frequency component. (iii) $\tau<0$,

$$\begin{aligned}I(\tau)\propto&\frac{(\xi_1-1)^2\exp(-2\alpha_2|\tau|)}{[\alpha_2^2+(\Delta_2^a)^2]^2}\\&+\frac{|\eta|^2(\xi_2-1)^2\exp(-2\alpha_3|\tau-\delta\tau|)}{[\alpha_3^2+(\Delta_3^a)^2]^2}+\exp(-\alpha_2|\tau|)\\&\times\exp(-\alpha_3|\tau-\delta\tau|)\{q'\exp[-i(\omega_3-\omega_2)\tau+i\omega_3\delta\tau]\\&+(q')^*\exp[i(\omega_3-\omega_2)\tau-i\omega_3\delta\tau]\},\end{aligned} \tag{14}$$

where

$$q'=\frac{\eta(\xi_1-1)(\xi_2-1)}{(\alpha_2-i\Delta_2^a)^2(\alpha_3-i\Delta_3^a)^2}.$$

Equation (14) is consistent with Eq. (5). Therefore, the requirement for the existence of a τ-dependent beat signal for $\tau<0$ is that the phase-correlated subpulses in beams 2 and 3 are overlapped temporally. Since beams 2 and 3 are mutually coherent, the temporal behavior of the beat signal should coincide with the case when the beams 2 and 3 are nearly monochromatic.

IV. PHOTON ECHO

It is interesting to understand the underlying physics in PBFS with incoherent lights. Much attention has been paid to the study of various ultrafast phenomena by using incoherent light sources recently [9,10]. For the phase matching condition $\vec{k}_2-\vec{k}_2'+\vec{k}_1$ and $\vec{k}_3-\vec{k}_3'+\vec{k}_1$ two sum-frequency trilevel echoes exist for the perturbation chain (I) and (II) [7]. Under the Doppler-broadened limit (i.e., $k_1u\to\infty$), if assuming that beam 2 (beam 3) has Gaussian line shape, then we have

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

$$\begin{aligned}P^{(\mathrm{I})}=&S_1(\vec{r})\exp[-i(\omega_1t+\omega_2\tau)]\\&\times\int_{-\infty}^{+\infty}d\vec{v}w(\vec{v})\int_0^\infty dt_3\int_0^\infty dt_2\int_0^\infty dt_1\exp[-i\theta_I(\vec{v})]\\&\times\exp[-(\Gamma_{10}+i\Delta_1)t_3]\exp[-(\Gamma_{20}+i\Delta_2+i\Delta_1)t_2]\\&\times\exp[-(\Gamma_{10}+i\Delta_1)t_1]\exp[-\beta_2^2(t_2-\tau)^2],\end{aligned} \tag{15}$$

$$\begin{aligned}P^{(\mathrm{II})}=&S_2(\vec{r})\exp[-i(\omega_1t+\omega_3\tau-\omega_3\delta\tau)]\\&\times\int_{-\infty}^{+\infty}d\vec{v}w(\vec{v})\int_0^\infty dt_3\int_0^\infty dt_2\int_0^\infty dt_1\exp[-i\theta_{\mathrm{II}}(\vec{v})]\\&\times\exp[-(\Gamma_{10}+i\Delta_1)t_3]\exp[-(\Gamma_{30}+i\Delta_3+i\Delta_1)t_2]\\&\times\exp[-(\Gamma_{10}+i\Delta_1)t_1]\exp[-\beta_3^2(t_2-\tau+\delta\tau)^2].\end{aligned} \tag{16}$$

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

$$\exp[-\beta_2^2(t_2-\tau)]\approx\frac{\sqrt{\pi}}{\beta_2}\delta(t_2-\tau), \tag{17}$$

$$\exp[-\beta_3^2(t_2-\tau+\delta\tau)]\approx\frac{\sqrt{\pi}}{\beta_3}\delta(t_2-\tau+\delta\tau). \tag{18}$$

When we substitute Eqs. (6), (7), (17), (18) into Eqs. (15), (16) we obtain (i) $\tau>\delta\tau$,

$$\begin{aligned}I(\tau)\propto&[(\xi_1-1)\tau/\beta_2]^2\exp(-2\Gamma_{20}^a|\tau|)\\&+|\eta|^2[(\xi_2-1)(\tau-\delta\tau)/\beta_3]^2\\&\times\exp(-2\Gamma_{30}^a|\tau-\delta\tau|)+[(\xi_1-1)(\xi_2-1)/\beta_2\beta_3]\tau\\&\times(\tau-\delta\tau)\exp(-\Gamma_{20}^a|\tau|)\exp(-\Gamma_{30}^a|\tau-\delta\tau|)\\&\times\{\eta\exp[-i(\Omega_3-\Omega_2)\tau-i(\xi_2-\xi_1)\Delta_1\tau\\&+i(\Omega_3+\xi_2\Delta_1)\delta\tau]+\eta^*\exp[i(\Omega_3-\Omega_2)\tau\\&+i(\xi_2-\xi_1)\Delta_1\tau-i(\Omega_3+\xi_2\Delta_1)\delta\tau]\}.\end{aligned} \tag{19}$$

This equation is consistent with Eq. (12). (ii) $0<\tau<\delta\tau$,

$$\frac{\sqrt{\pi}}{\beta_3}\delta(t_2-\tau+\delta\tau)=0.$$

In this case, photon echo does not exist for the perturbation chain (II). Then $I(\tau)\propto[(\xi_1-1)\tau/\beta_2]^2\exp(-2\Gamma_{20}^a\tau)$, this equation is consistent with Eq. (13). (iii) $\tau<0$,

$$\frac{\sqrt{\pi}}{\beta_2}\delta(t_2-\tau)=0.$$

In this case, photon echo does not exist for the perturbation chains (I) and (II). This case is consistent with Eq. (5).

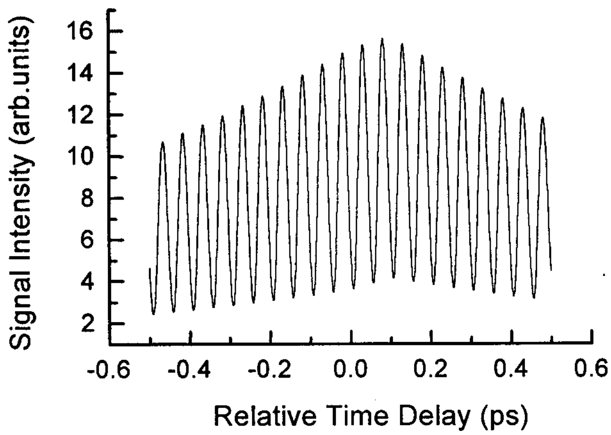

FIG. 3. Beat signal intensity versus relative time delay.

V. EXPERIMENT AND RESULT

We performed the PBFS in sodium vapor, where the ground state $3S_{1/2}$, the intermediate state $3P_{3/2}$, and two excited states $6S_{1/2}$ and $5D_{3/2,5/2}$ formed 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 ω_1, ω_2, and ω_3. DL1, DL2, and DL3 had linewidth 0.1 nm and pulse width 5 ns. DL1 was tuned to 589.0 nm, the wavelength of the $3S_{1/2}$-$3P_{3/2}$ transition; DL2 was tuned to 515.4 nm, the wavelength of the $3P_{3/2}$-$6S_{1/2}$ transition; while DL3 was tuned to 498.3 nm, the wavelength of the $3P_{3/2}$-$5D_{3/2,5/2}$ transition. A beam splitter was used to combine the ω_2 and ω_3 components derived from DL2 and DL3, respectively, for beams 2 and 3, which intersected in the oven containing the Na vapor. The time delay τ between beams 2 and 3 could be varied by an optical delay line. Beam 1, which propagated along a direction opposite to 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 to that of beam 3. It was detected by a photodiode.

We measured the beat signal intensity as a function of the time delay between beams 2 and 3. Figure 3 presents the result. It shows that as τ varied, the beat signal intensity modulates sinusoidally with period 50 fs. The modulation frequency can be obtained more directly by making a Fourier transformation of the PBFS data. Figure 4 presents the Fourier spectrum of the data in which τ is varied for a range of 15 ps. Then we obtain the modulation frequency 126 ps^{-1} corresponding to the beating between the resonant frequencies of the transitions from $3P_{3/2}$ to $6S_{1/2}$ and from $3P_{3/2}$ to $5D_{3/2,5/2}$. On the other hand, the temporal behavior of the beat signal is quite asymmetric with the maximum of the signal shifted from $\tau=0$. We attribute this asymmetry to the difference in the zero time delay between beams 2 and 3 for the ω_2 and ω_3 frequency components. To confirm this, we measured the τ dependence of the FWM signal when beams 2 and 3 only consisted of only one frequency component. Figures 5 and 6 present the results when the frequencies of beams 2 and 3 are ω_2 and ω_3, respectively. The difference in the zero-time delay is obvious in these figures. It is due to the large difference between the wavelengths of DL2 and DL3 so that the dispersion of the optical components becomes important. This can be understood as follows. Con-

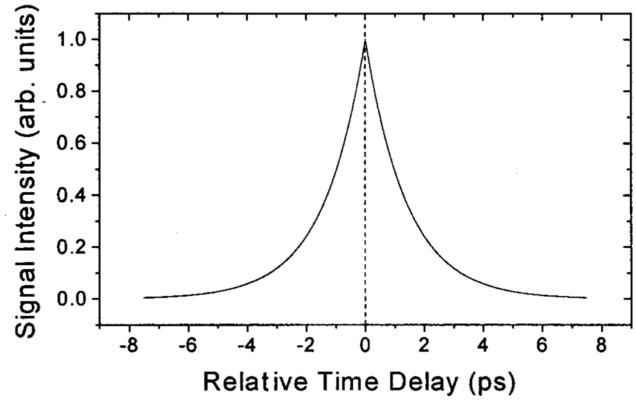

FIG. 5. FWM signal intensity versus relative time delay when pump beams consist of only ω_2.

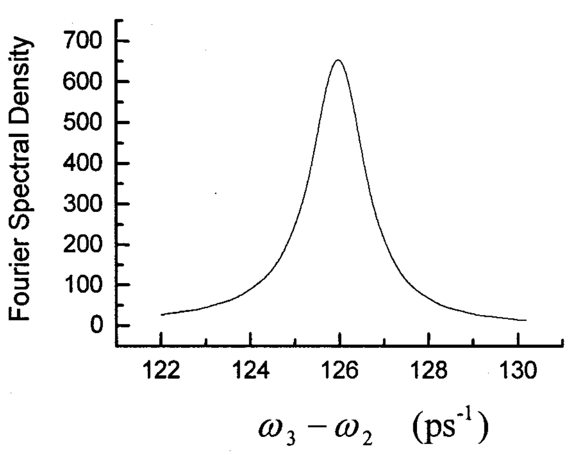

FIG. 4. Fourier spectrum of the experimental data in which τ is varied for a range of 15 ps.

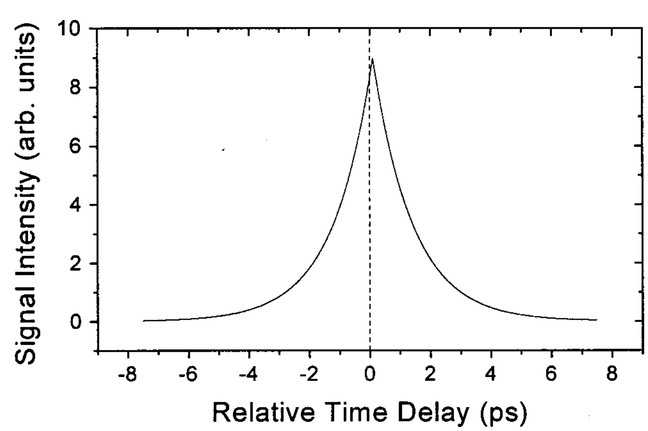

FIG. 6. FWM signal intensity versus relative time delay when pump beams consist of only ω_3.

sider the case that the optical paths between 2 and 3 are equal for the ω_2 component. Owing to the difference between the zero time delays for the ω_2 and ω_3 frequency components, the optical paths between beams 2 and 3 will be different by $c\delta\tau$ for the ω_3 component. As the result, there is an extra phase factor $\omega_3\delta\tau$ for the ω_3 frequency component. The difference between the zero time delays for the ω_2 and ω_3 frequency components corresponds to the propagation of beams in the glass (mainly the prism in the optical delay line) for a distance [4,5].

PBFS can be considered as a technique, which possesses the main features of the laser spectroscopies in the frequency domain and in the time domain. First PBFS is closely related to the Doppler-free two-photon absorption spectroscopy in tuning ω_2 and ω_3 to the resonant frequencies when narrow-band lights are used. However unlike the techniques in the frequency domain, here we are interested in the temporal behavior of the signal and the frequencies of the laser do not need be calibrated. In this sense PBFS is similar to the spectroscopies in the time domains. PBFS is related intrinsically to the sum-frequency trilevel photon echo when broadband lights are used. In this case, when pulse laser beams 2 and 3 are separated temporally, then before the application of beam 3 the polarization exhibits free evolution. As a result, the modulation frequency is directly related to the energy level of the system no matter whether the beams have a narrow-band or broadband linewidth. The advantage of PBFS over other time-domain techniques is that the temporal resolution is not limited by the laser pulse width.

In conclusion, we have employed a second-order coherence function theory to study the effect of laser coherence on polarization beats in four-level system. It is found that, the temporal behavior of the beat signal depends on the stochastic properties of the lasers and transverse relaxation rate of the atomic energy-level system. We have considered the cases that pump beams have either narrow band or broad-band linewidth and found that for both cases a Doppler-free precision in the measurement of the energy-level splitting between two excited states which are dipolar forbidden from the ground state can be achieved. We also studied the asymmetric behavior of the polarization beats, we attribute this asymmetry to the shift of the zero time delay between beams 2 and 3 for the ω_2 and ω_3 frequency components which is due to the dispersion of the optical components. It is worth mentioning that the asymmetric behavior of the polarization beat signal in a four-level system do not affect the overall accuracy of using PBFS to measure the energy-level splitting. Furthermore PBFS can tolerate small perturbations of the optical path due to mechanical vibration and distortion of the optical components as long as these are small compared with $c/|\omega_3-\omega_2|$.

ACKNOWLEDGMENT

The authors gratefully acknowledge financial support from the Chinese National Nature Sciences Foundation (Grant No. 69978019).

[1] D. DeBeer, L. G. Van Wagenen, R. Beach, and S. R. Hartmann, Phys. Rev. Lett. **56**, 1128 (1986).
[2] P. M. Fu, Z. H. Yu, X. Mi, X. F. Li, and Q. Jiang, Phys. Rev. A **50**, 698 (1994).
[3] P. M. Fu, X. Mi, Z. H. Yu, Q. Jiang, Y. P. Zhang, and X. F. Li, Phys. Rev. A **52**, 4867 (1995).
[4] R. Beach, D. DeBeer, and S. R. Hartmann, Phys. Rev. A **32**, 3467 (1985).
[5] X. Mi, Z. H. Yu, Q. Jiang, and P. M. Fu, Opt. Commun. **116**, 443 (1995).
[6] J. C. Kirkwood, D. J. Ulness, and A. C. Albrecht, J. Chem. Phys. **108**, 9425 (1998).
[7] T. W. Mossberg, R. Kachru, S. R. Hartmann, and A. M. Flusberg, Phys. Rev. A **20**, 1976 (1979).
[8] B. Picinbono and E. Boileau, J. Opt. Soc. Am. **58**, 784 (1968).
[9] N. Morita and T. Yajima, Phys. Rev. A **30**, 2525 (1984).
[10] M. Mitsunaga and R. G. Brewer, Phys. Rev. A **32**, 1605 (1985).