Volume 9, Number 12
1009-1963/2000/09(12)/0905-05
CHINESE PHYSICS
December, 2000
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RAMAN-ENHANCED POLARIZATION BEATS WITH PHASE-CONJUGATION GEOMETRY*

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(Received 27 April 1999; revised manuscript received 1 July 1999)

The Raman-enhanced polarization beats (REPB) with phase-conjugation geometry in carbon disulfide are investigated using chaotic and phase-diffusion models. The overall accuracy of using REPB to measure the resonant frequency of Raman-active vibrational mode is determined by the relaxation rates of the Raman mode and the molecular-reorientational grating. Different stochastic models of the laser field only affect the sixth- and fourth-order coherence function. The difference between the REPB and the fourth-order coherence on ultrafast modulation spectroscopy is discussed as well from a physical viewpoint.

Keywords: Raman-enhanced Polarization Beat, Sixth-order coherence, Phase and amplitude fluctuations
PACC: 4265, 4250, 3290

I. INTRODUCTION

Polarization beats, which originate from the interference between the macroscopic polarizations, have attracted a lot of attention recently.[1−9] It is closely related to quantum beats, Raman quantum beats,[1] or FOCUMS.[7] The atomic response to stochastic optical fields is now largely well understood. There exist methods to calculate the atomic density matrix elements for a wide variety of field statistics, including phase-diffusing fields, phase-diffusing fields with colored noise, chaotic fields, real Gaussian fields, and phase jump fields.[3,7,9] The chaotic field model and the Brownian-motion phase-diffusion model are considered in parallel with a discussion on Raman-enhanced polarization beat (REPB). We develop a unified theory which involves sixth-order coherence-function to study the influence of partial-coherence properties of pump beams on polarization beats. Our sixth-order coherence function theory includes both the finite light bandwidth effect and constant background contribution. This is of vital importance in the REPB.

Ⅱ. BASIC THEORY

REPB is a third-order nonlinear polarization beat phenomenon. The basic geometry is shown in Fig.1. Beams 1 and 2 consist of two frequency components ω_1 and ω_3, a small angle exists between them. Beam 3 with frequency ω_3 is almost propagating along the opposite direction of beam 1. In a Kerr medium, the nonlinear interaction of beams 1 and 2 with the medium gives rise to two molecular-reorientational gratings, i.e., ω_1 and ω_3 will induce their own static gratings G1 and G2, respectively. The four-wave mixing(FWM) signal is the result of the diffraction of beam 3 by G1 and G2.

We assume that ω_1 and ω_3 are far off resonance with electronic transitions. If $|\omega_1 - \omega_3|$ is near the Raman resonant frequency Ω_R, a large angle moving grating and two small angle moving gratings, which are formed by the interference of beams 2, 3 and beams 1, 2, respectively, will excite the Raman-active vibrational mode of the medium and enhance the FWM signal. The beat signal (beam 4) is along the opposite direction of beam 2 approximately.

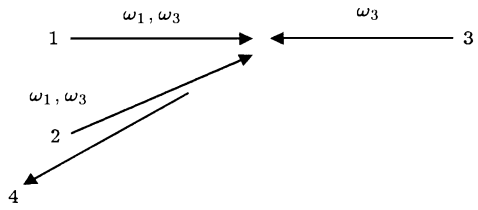


Fig.1. Schematic diagram of the geometry of REPB.

A THE CHAOTIC FIELD

The FWM signal is proportional to the average of the absolute square of the nonlinear polarization $P^{(3)}$ over the random variables of the stochastic process

*Supported by the National Natural Science Foundation of China (Grant No. 69978019).

$I(\Delta,\tau) \propto < |P^{(3)}|^2 >$, which involves sixth-, fourth- and second-order coherence functions of $u_i(t)$ in phase conjugation geometry. $u_i(t)$ is a dimensionless statistical factor that contains phase and amplitude fluctuations. 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 fluctuations of both the amplitude and the phase of the field. $u_i(t)$ has Gaussian statistics, with its sixth- and fourth-order coherence function satisfying[9]

$$\begin{aligned} &< u_i(t_1)u_i(t_2)u_i(t_3)u_i^*(t_4)u_i^*(t_5)u_i^*(t_6) > \\ =&< u_i(t_1)u_i^*(t_4) >< u_i(t_2)u_i(t_3)u_i^*(t_5)u_i^*(t_6) > \\ &+ u_i(t_1)u_i^*(t_5) >< u_i(t_2)u_i(t_3)u_i^*(t_4)u_i^*(t_6) > \\ &+ < u_i(t_1)u_i^*(t_6) >< u_i(t_2)u_i(t_3)u_i^*(t_4)u_i^*(t_5) >, \end{aligned} \tag{1}$$

and

$$\begin{aligned} &< u_i(t_1)u_i(t_2)u_i^*(t_3)u_i^*(t_4) >=< u_i(t_1)u_i^*(t_3) > \\ &< u_i(t_2)u_i^*(t_4) > + < u_i(t_1)u_i^*(t_4) >< u_i(t_2)u_i^*(t_3) > . \end{aligned} \tag{2}$$

Further assuming that the laser sources have Lorentzian line shape, we have

$$< u_i(t_1)u_i^*(t_2) >= \exp(-\alpha_i|t_1-t_2|). \tag{3}$$

Here $\alpha_i = \frac{1}{2}\delta\omega_i, \delta\omega_i$, is the linewidth of the laser with frequency ω_i.

We first consider the situation when the laser linewidths of beams 1, 2 and 3 are broadband (i.e., $\alpha_1, \alpha_3 >> \gamma, \gamma_R$). In this limit, after performing the tedious integration we obtain for (i) $\tau > 0$

$$\begin{aligned} &I(\Delta,\tau) \propto (1+\eta_1^2+\eta_2^2)\frac{\chi_R^2\gamma_R(\alpha_1+\alpha_3)}{(\alpha_1+\alpha_3)^2+\Delta^2} \\ &-\chi_R\chi\gamma_R\gamma\frac{\Delta[(5\alpha_1+\alpha_3)(\alpha_1+\alpha_3)+\Delta^2]}{\alpha_1[(\alpha_1+\alpha_3)^2+\Delta^2]^2} \\ &+\frac{\chi^2\gamma}{2\alpha_1\alpha_3}(\alpha_1\eta_1^2\eta_2^2+\alpha_3)+\left\{\frac{\chi_R^2\gamma_R^2(\alpha_1+\alpha_3)}{\alpha_1[(\alpha_1+\alpha_3)^2+\Delta^2]}\right. \\ &\left.-\frac{2\chi_R\chi\gamma_R\Delta}{(\alpha_1+\alpha_3)^2+\Delta^2}+\chi^2\right\}\exp(-2\alpha_1|\tau|) \\ &+\eta_2^2\left\{\frac{\chi_R^2\gamma_R^2(\alpha_1+\alpha_3)}{\alpha_3[(\alpha_1+\alpha_3)^2+\Delta^2]}+\chi^2\eta_1^2\right\}\exp(-2\alpha_3|\tau|) \\ &+4\eta_1\eta_2\exp[-(\alpha_1+\alpha_3)|\tau|]\left\{\left[\frac{\chi^2}{2}-\frac{\chi\chi_R\gamma_R\Delta}{2[(\alpha_1+\alpha_3)^2+\Delta^2]}\right]\right. \\ &\times\cos[\Delta\boldsymbol{k}\cdot\boldsymbol{r}-(\omega_1-\omega_3)\tau] \\ &\left.-\frac{\chi\chi_R\gamma_R(\alpha_1+\alpha_3)}{2[(\alpha_1+\alpha_3)^2+\Delta^2]}\sin[\Delta\boldsymbol{k}\cdot\boldsymbol{r}-(\omega_1-\omega_3)\tau]\right\}, \end{aligned} \tag{4}$$

and (ii) $\tau < 0$

$$\begin{aligned} &I(\Delta,\tau) \propto (1+\eta_1^2+\eta_2^2)\frac{\chi_R^2\gamma_R(\alpha_1+\alpha_3)}{(\alpha_1+\alpha_3)^2+\Delta^2} \\ &-\chi_R\chi\gamma_R\gamma\frac{\Delta[(5\alpha_1+\alpha_3)(\alpha_1+\alpha_3)+\Delta^2]}{\alpha_1[(\alpha_1+\alpha_3)^2+\Delta^2]^2} \\ &+\frac{\chi^2\gamma}{2\alpha_1\alpha_3}(\alpha_1\eta_1^2\eta_2^2+\alpha_3)+\frac{3\chi^2\eta_1^2\eta_2^2\gamma^2}{2\alpha_3^2}\exp(-2\gamma|\tau|) \\ &+2\chi_R^2\gamma_R^2\left\{\frac{\alpha_1\eta_2^2+\alpha_3}{(\alpha_1+\alpha_3)^2+\Delta^2}\right. \\ &\left.+\frac{(1+\eta_2^2)[(\alpha_1+\alpha_3)^2-\Delta^2]}{[(\alpha_1+\alpha_3)^2+\Delta^2]^2}\right\}\exp(-2\gamma_R|\tau|) \\ &+\exp[-(\gamma_R+\gamma)|\tau|]\frac{4\eta_1\eta_2\chi\chi_R\gamma\gamma_R}{\alpha_3[(\alpha_1+\alpha_3)^2+\Delta^2]^2} \\ &\times\{(\alpha_1+\alpha_3)(2\gamma_R+\gamma)\Delta\cos \\ &\times[\Delta\boldsymbol{k}\cdot\boldsymbol{r}-(\omega_1-\omega_3)\tau+\Delta|\tau|] \\ &-[(\alpha_1+2\alpha_3)(\alpha_1+\alpha_3)^2+\alpha_1\Delta^2]\sin \\ &\times[\Delta\boldsymbol{k}\cdot\boldsymbol{r}-(\omega_1-\omega_3)\tau+\Delta|\tau|]\} \end{aligned} \tag{5}$$

Here, $\eta_1 = \frac{\varepsilon_2}{\varepsilon_1}$, $\eta_2 = \frac{\varepsilon_2'}{\varepsilon_1'}$; $\Delta = |\omega_1-\omega_3| - \Omega_R$; $\Delta\boldsymbol{k} = (\boldsymbol{k}_1-\boldsymbol{k}_1') - (\boldsymbol{k}_2-\boldsymbol{k}_2')$; γ and $\chi(\gamma_R, \chi_R)$ are the relaxation rate and the nonlinear susceptibility of the two static gratings (the Raman mode), respectively; ε_i and $\boldsymbol{k}_i(\varepsilon_i', \boldsymbol{k}_i')$ are the constant field amplitude and the wave vector of frequency components ω_1 and ω_3 in beam 1 (beam 2), respectively; ε_3 and $\boldsymbol{k}_3$ are the field amplitude and the wave vector of beam 3, respectively; τ is the time delay of beam 2 with respect to beam 1.

Equation (4) indicates that when $\tau > 0$, the temporal behavior of the beat signal intensity reflects mainly the characteristic of the lasers, i.e., the frequency $\omega_3 - \omega_1$ and the damping rate $\alpha_1 + \alpha_3$ of the modulation are determined by the incident laser beams. If we employ REPB to measure the modulation frequency $\omega_d = \omega_3 - \omega_1$, the accuracy can be improved by measuring as many cycles of the modulation as possible. Since the amplitude of the modulation decays with a time constant $(\alpha_1+\alpha_3)^{-1}$ as $|\tau|$ increases, the maximum $|\tau|$ should equal approximately $2(\alpha_1+\alpha_3)^{-1}$. We obtain the theoretical limit of the uncertainty of the modulation frequency measurement $\Delta\omega_d$, which is $\Delta\omega_d \approx \pi(\alpha_1+\alpha_3)$, i.e., in the modulation frequency measurement the theoretical limit of the accuracy is related to the decay time constant of the beat signal modulation amplitude. In this case, the precision of using REPB to measure the resonant frequencies of the Raman mode is determined by how well $\omega_3-\omega_1$ can be tuned to Ω_R. On the other hand, when $\tau < 0$, Eq.(5) indicates that the beat sig-

nal modulates with a frequency $(\omega_3-\omega_1)-\Delta=\Omega_R$ and a damping rate $\gamma_R+\gamma$ as τ is varied. We can obtain the resonant frequencies of the Raman vibrational mode with an accuracy given by $\pi(\gamma_R+\gamma)$ approximately, which is determined by material property.

Different stochastic models of the laser field affect only the sixth- and fourth-order coherence function. The constant terms in relations (4) and (5), which are independent of the relative time-delay between beam 1 and beam 2, originate from the phase fluctuation of the chaotic field. The modulation terms including the factors $\exp[-(\alpha_1+\alpha_3)|\tau|]$, or $\exp[-\gamma_R+\gamma)|\tau|]$ also originate from the phase fluctuation of the chaotic field. In contrast, the decay terms including the factors $\exp(-2\alpha_1|\tau|)$, $\exp(-2\alpha_3|\tau|)$, $\exp(-\gamma|\tau|)$, or $\exp(-2\gamma_R|\tau|)$ originate from the amplitude fluctuation of the chaotic field. On the other hand, REPB is different for $\tau>0$ and $\tau<0$. However, as $|\tau|\to\infty$, Eq.(4) is identical to Eq.(5). Physically, when $|\tau|\to\infty$, beams 1 and 2 are mutually incoherent, therefore whether τ is positive or negative does not affect the REPB.

We then consider the situation when the laser linewidths of beams 1, 2 and 3 are narrow band (i.e., $\alpha_1,\alpha_3<<\gamma,\gamma_R$). In this limit, after performing the tedious integration we obtain

$$
\begin{aligned}
&I(\Delta,\tau)\propto(1+2\eta_1^2+\eta_2^2)\frac{\chi_R^2\gamma_R^2}{\gamma_R^2+\Delta^2}\\
&-\frac{2\chi_R\chi\gamma_R\Delta}{\gamma_R^2+\Delta^2}+\chi^2(1+2\eta_1^2\eta_2^2)\\
&+\left(\frac{\chi_R^2\gamma_R^2}{\gamma_R^2+\Delta^2}-\frac{2\chi_R\chi\gamma_R\Delta}{\gamma_R^2+\Delta^2}+\chi^2\right)\exp(-2\alpha_1|\tau|)\\
&+\eta_2^2\left(\frac{\chi_R^2\gamma_R^2}{\gamma_R^2+\Delta^2}+4\eta_1^2\chi^2\right)\exp(-2\alpha_3|\tau|)\\
&+4\eta_1\eta_2\exp[-(\alpha_1+\alpha_3)|\tau|]\Bigg\{\left(\chi^2-\frac{\chi\chi_R\gamma_R\Delta}{\gamma_R^2+\Delta^2}\right)\\
&\cdot\cos[\Delta\boldsymbol{k}\cdot\boldsymbol{r}-(\omega_1-\omega_3)\tau]\\
&-\chi\chi_R\gamma_R\frac{\gamma_R(\gamma_R+\gamma)^2+\Delta^2(\gamma_R+2\gamma)}{(\gamma_R^2+\Delta^2)[(\gamma_R+\gamma)^2+\Delta^2]}\\
&\cdot\sin[\Delta\boldsymbol{k}\cdot\boldsymbol{r}-(\omega_1-\omega_3)\tau]\Bigg\},
\end{aligned}
\tag{6}
$$

This case when the pump beams have a narrow-band linewidth is similar to Eq.(4).

B THE PHASE-DIFFUSION FIELD

We have assumed that the laser sources are chaotic field in the above calculation. Another commonly used stochastic model is 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 sixth-and fourth-order coherence functions are[7]

$$
\begin{aligned}
&<u_i(t_1)u_i(t_2)u_i(t_3)u_i^*(t_4)u_i^*(t_5)u_i^*(t_6)>\\
&=\exp[-\alpha_i(|t_1-t_4|+|t_1-t_5|+|t_1-t_6|+|t_2-t_4|\\
&+|t_2-t_5|+|t_2-t_6|\\
&+|t_3-t_4|+|t_3-t_5|+|t_3-t_6|)]\\
&\times\exp[\alpha_i(|t_1-t_2|+|t_1-t_3|+|t_2-t_3|\\
&+|t_4-t_5|+|t_4-t_6|+|t_5-t_6|)],
\end{aligned}
\tag{7}
$$

and

$$
\begin{aligned}
&<u_i(t_1)u_i(t_2)u_i^*(t_3)u_i^*(t_4)>\\
&=\exp[-\alpha_i(|t_1-t_3|+|t_1-t_4|+|t_2-t_3|+|t_2-t_4|)]\\
&\times\exp[\alpha_i(|t_1-t_2|+|t_3-t_4|)].
\end{aligned}
\tag{8}
$$

We first consider the situation when the laser linewidths of beams 1, 2 and 3 are broadband (i.e., $\alpha_1,\alpha_3>>\gamma,\gamma_R$). (i) For $\tau>0$, we have

$$
\begin{aligned}
&I(\Delta,\tau)\propto(1+\eta_1^2+\eta_2^2)\frac{\chi_R^2\gamma_R(\alpha_1+\alpha_3)}{(\alpha_1+\alpha_3)^2+\Delta^2}\\
&-\chi_R\chi\gamma_R\gamma\frac{\Delta[(5\alpha_1+\alpha_3)(\alpha_1+\alpha_3)+\Delta^2]}{2\alpha_1[(\alpha_1+\alpha_3)^2+\Delta^2]^2}\\
&+\frac{\chi^2\gamma}{2\alpha_1\alpha_3}(\alpha_1\eta_1^2\eta_2^2+\alpha_3)+2\eta_1\eta_2\exp[-(\alpha_1+\alpha_3)|\tau|]\\
&\times\Bigg\{\left[\chi^2-\frac{\chi\chi_R\gamma_R\Delta}{(\alpha_1+\alpha_3)^2+\Delta^2}\right]\cos[\Delta\boldsymbol{k}\cdot\boldsymbol{r}-(\omega_1-\omega_3)\tau]\\
&-\frac{\chi\chi_R\gamma_R(\alpha_1+\alpha_3)}{(\alpha_1+\alpha_3)^2+\Delta^2}\sin[\Delta\boldsymbol{k}\cdot\boldsymbol{r}-(\omega_1-\omega_3)\tau]\Bigg\},
\end{aligned}
\tag{9}
$$

and (ii) for $\tau<0$, we have

$$
\begin{aligned}
&I(\Delta,\tau)\propto(1+\eta_1^2+\eta_2^2)\frac{\chi_R^2\gamma_R(\alpha_1+\alpha_3)}{(\alpha_1+\alpha_3)^2+\Delta^2}\\
&-\chi_R\chi\gamma_R\gamma\frac{\Delta[(5\alpha_1+\alpha_3)(\alpha_1+\alpha_3)+\Delta^2]}{2\alpha_1[(\alpha_1+\alpha_3)^2+\Delta^2]^2}\\
&+\frac{\chi^2\gamma}{2\alpha_1\alpha_3}(\alpha_1\eta_1^2\eta_2^2+\alpha_3)-\exp[-(\gamma_R+\gamma)|\tau|]\\
&\times\frac{4\eta_1\eta_2\chi\chi_R\gamma\gamma_R[(\alpha_1+2\alpha_3)(\alpha_1+\alpha_3)^2+\alpha_1\Delta^2]}{\alpha_3[(\alpha_1+\alpha_3)^2+\Delta^2]}\\
&\times\sin[\Delta\boldsymbol{k}\cdot\boldsymbol{r}-(\omega_1-\omega_3)\tau+\Delta|\tau|],
\end{aligned}
\tag{10}
$$

Equations (9) and (10) are different from the result based on a chaotic model. The constant terms in Eqs.(9) and (10), which are independent of τ, originate from the phase fluctuations of the phase-diffusion field. The modulation terms including the factors $\exp[-(\alpha_1+\alpha_3)|\tau|]$, or $\exp[-(\gamma_R+\gamma)|\tau|]$ also originate from the phase fluctuation of the phase-diffusion field. But Eqs.(9) and (10) are short of

the decay terms including the factors $\exp(-2\alpha_1|\tau|)$, $\exp(-2\alpha_3|\tau|)$, $\exp(-2\gamma|\tau|)$, or $\exp(-\gamma_R|\tau|)$. The drastic difference of the results also exists in the fourth order coherence on ultrafast modulation spectroscopy (FOCUMS) when these two models are employed.[7] Physically, the chaotic field has the property of photon bunching, which can affect any multiphoton process when the higher-order correlation function of the field plays an important role.

We then consider the situation when the laser linewidths of beams 1, 2 and 3 are narrow band (i.e., $\alpha_1, \alpha_3 << \gamma, \gamma_R$). In this limit, after performing the tedious integration we obtain

$$
\begin{aligned}
&I(\Delta,\tau) \propto (1+\eta_1^2+\eta_2^2)\frac{\chi_R^2\gamma_R^2}{\gamma_R^2+\Delta^2} \\
&-\frac{\chi_R\chi\gamma_R\Delta}{\gamma_R^2+\Delta^2}+\chi^2(1+\eta_1^2\eta_2^2) \\
&+2\eta_1\eta_2\exp[-(\alpha_1+\alpha_3)|\tau|] \\
&\times\Bigg\{\Big(\chi^2-\frac{\chi_R\chi\gamma_R\Delta}{\gamma_R^2+\Delta^2}\Big)\cos[\Delta\boldsymbol{k}\cdot\boldsymbol{r}-(\omega_1-\omega_3)\tau] \\
&-\frac{\chi_R\chi\gamma_R^2}{\gamma_R^2+\Delta^2}\Big)\sin[\Delta\boldsymbol{k}\cdot\boldsymbol{r}-(\omega_1-\omega_3)\tau]\Bigg\} \qquad (11)
\end{aligned}
$$

Equation (11) is analogous to (9), which indicates that the temporal behavior of the beat signal intensity reflects mainly the characteristic of the lasers.

The main purpose of the above discussion is that we reveal an important fact that the amplitude fluctuation of the laser field plays a critical role in the temporal behavior of REPB signal. Furthermore, the different roles of the phase fluctuation and amplitude fluctuation have been pointed out in the time-domain. This is quite different from the time-delayed FWM with incoherent light in a two-level system,[10] where T_2 of the system can be deduced. For the latter case, the phase fluctuation of the light field is crucial. The REPB is also different from the FOCUMS. The phase fluctuation of the light field is crucial in the FOCUMS. On the other hand, because of $< u_i(t) >= 0$ and $< u_i^*(t) >= 0$, the absolute square of the stochastic average of the polarization $| < P^{(3)} > |^2$, which involves second-order coherence function of $u_i(t)$, cannot be used to describe the temporal behavior of the REPB.[8] Therefore, the sixth-order coherence function theory of chaotic field is of vital importance in REPB.

Ⅲ. EXPERIMENT AND RESULT

We are interested in the temporal behavior of the REPB signal intensity with fixed frequency detuning. The carbon disulfide (CS_2) with 655.7-cm^{-1} vibrational mode was contained in a sample cell with a thickness of 9 mm. The second harmonic of a Quanta-Ray YAG laser was used to pump two dye lasers (DL1 and DL2). DL1 and DL2 had linewidth 0.05nm, pulse width 5ns and output energy 1 mJ. The first dye laser DL1 had a wavelength of 589nm. The wavelength of DL2 was approximately 567nm and could be scanned by a computer-controlled stepping motor. A beam splitter was used to combine the ω_1 and ω_3 components derived from DL1 and DL2, respectively, for beams 1 and 2, which intersected in the sample with a small angle $\theta = 1.5°$. The relative time τ between beams 1 and 2 could be varied. Beam 3, which propagated along the direction opposite to that of beam 1, was derived from DL2. 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 2. It was detected by a photodiode.

We first performed a nondegenerate FWM experiment in which beams 1 and 2 only consisted of ω_1 frequency component. We measured the REN-FWM spectrum with a fixed time delay (at τ=0 ps) by scanning ω_3. Our results are shown in Fig.2, which showed an asymmetrical resonant profile due to the interference between the Raman resonant term and the nonresonant background originating solely from the molecular reorientational grating.[11] From this spectrum ω_3 was tuned to the resonant frequency (i.e., $\Delta = |\omega_1-\omega_3|-\Omega_R$=0). We then performed the REPB experiment with fixed frequency detuning by measuring the beat signal intensity as a function of the relative time delay when beams 1 and 2 consist of both frequencies ω_1 and ω_3. Figure 3 presents the result of the beat experiment. The beat signal intensity modulates sinusoidally with a period of 50.5 fs. The modulation frequency can be obtained more directly by making a Fourier transformation of the REPB data. Figure 4 shows the Fourier spectrum of the REPB data, in which τ is varied for a range of 5 ps. Then we obtain the modulation frequency $1.24\times10^{14}\,\mathrm{sec}^{-1}$, corresponding to the resonant frequency of the Raman vibrational mode.

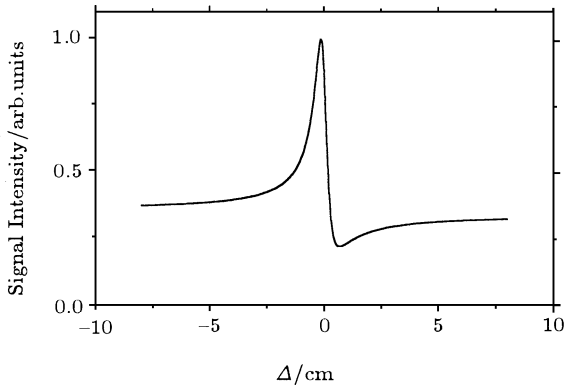


Fig.2. RENFWM spectrum at τ=0 ps.

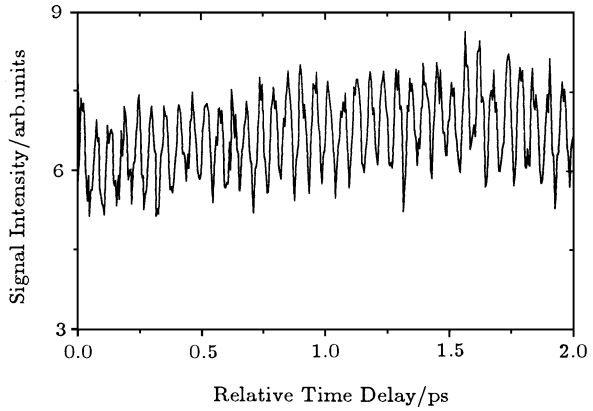


Fig.3. Experimental result of the beat signal intensity versus relative time delay.

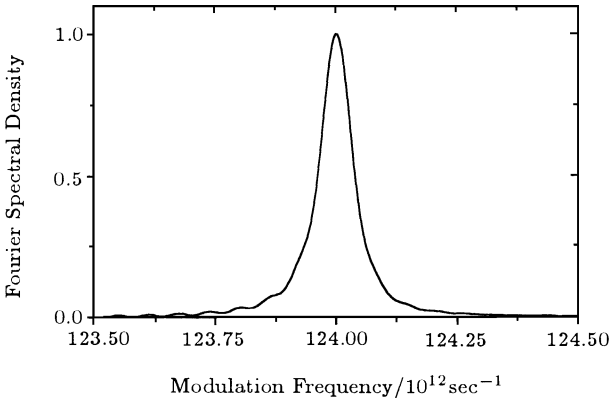


Fig.4. Fourier spectrum of the REPB data in which τ is varied for a range of 5 ps.

In conclusion, we have employed a sixth-order coherence function theory to study the Raman-enhanced polarization beats with phase-conjugation geometry. The measurement accuracy for the resonant frequency of Raman-active vibrational mode is related to the decay time constant of the beat signal modulation amplitude. We also show that the temporal behavior of the REPB signal is different drastically if the laser sources are described by a phase-diffusion model.

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