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NONLINEAR EFFECTS OF THIRD-AND FIFTH-ORDER ON POLARIZATION BEATS IN A FOUR-LEVEL SYSTEM*

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We have employed second-order coherence function theory to study the nonlinear effects of third-and fifth-order on polarization beats in a four-level system (TPBFS and FPBFS). It is found that the different temporal behavior of the beat signal in TPBFS and FPBFS depends on the stochastic properties of the lasers and transverse relaxation rate of the transition. We have considered the cases that pump beams have either narrow band or broadband linewidth and found that for both cases a Doppler-free precision can be achieved in the measurement of the energy-level difference between two excited states which are dipolar forbidden from the ground state. We also discussed the spatiall modulation behavior of the beat signal.

Keywords: nonlinear effect, polarization beat, wave-mixing
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I. INTRODUCTION

Polarization beats, which originate from the interference between the macroscopic polarizations, have attracted a lot of attention recently.[1−4] 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[5,6] and cascade three-level systems.[7] In the quasi-two-level, the excited and the ground states consist of sub-level structures. Quantum beat manifests itself as an oscillation of the signal with frequency corresponding to the energy-level splittings. For example, DeBeer *et al.*[5] performed the first ultrafast modulation spectroscopy (UMS) experiment in sodium vapor. The beating signal exhibits 1.9ps modulation corresponding to the sodium D-line splitting when the time delay between two double-frequency pump beams increases. Fu *et al.*[6] 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, UMS technique has also been applied to a cascade three-level system.[7] Ultrafast modulation spectroscopy in a 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 sates is well known, then from the beating the energy separation between the intermediate and excited states can be deduced. These phenomena result from the third-order polarization interference between two different channels. Ma *et al.*[8,9] developed a polarization beat interferometry to measure the phase of the third-and fifth-order susceptibilities of the Cd(S, Se)-doped glasses. They also observed interference between the third-and the fifth-order polarization in semiconductor doped glasses.[10] Based on the interference between two two-photon processes, we studied the UMS technique in four-level system.

In this paper, we report the nonlinear effects of third-and fifth-order on polarization beats in 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 phase-diffusion model, which is used to describe an

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amplitude-stabilized laser source. This model assumes that the amplitude of the laser field is 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 nonlinear effects of third-and fifth-order on polarization beats in a four-level system. If TPBFS and FPBFS are 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.

Ⅱ. BASIC THEORY

UMS in a four-level system 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 exicted 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 an $\Omega_2(\Omega_3)$, respectively, while states between $|2\rangle$ and $|3\rangle$ and between $|0\rangle$ and $|\rangle 2(|3\rangle)$ are dipolar forbidden. We consider in this four-level system a double-frequency time-delay four-wave-mixing (FWM) and six-wave mixing (SWM) polarization beats 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). All the incident beams were linearly polarized in the same direction. 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 third-order polarization beats, 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. Two two-photon FWM signals (beam 4) with frequency ω_1 will be emitted along the directions $\boldsymbol{k}_2 - \boldsymbol{k}_2' + \boldsymbol{k}_1$ and $\boldsymbol{k}_3 - \boldsymbol{k}_3' + \boldsymbol{k}_1$, respectively. In fifth-order polarization beats, three two-photon nondegenerate SWM signals (beam 5) with frequency ω_1 will be emitted along the directions $\boldsymbol{k}_1 + 2\boldsymbol{k}_2 - 2\boldsymbol{k}_2', \boldsymbol{k}_1 + \boldsymbol{k}_2 - \boldsymbol{k}_2' + \boldsymbol{k}_3 - \boldsymbol{k}_3'$ and $\boldsymbol{k}_1 + 2\boldsymbol{k}_3 - 2\boldsymbol{k}_3'$. In the phase-conjugation geometry, beams 4 and 5 have a small angle between them.

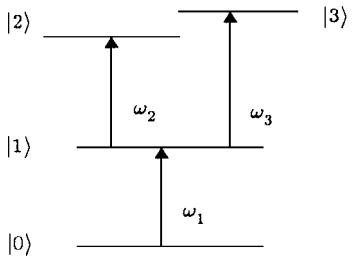


Fig.1. Four-level configuration to be treated in TPBFS and FPBFS.

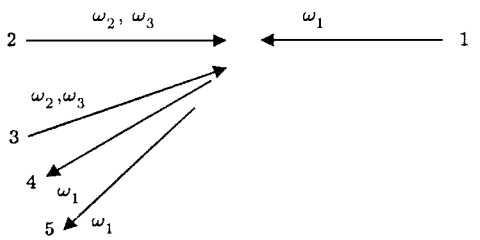


Fig.2. Schematic diagram of the geometry of TPBFS and FPBFS.

The third-order off-diagonal density matrix element $\rho_{10}^{(3)}$, which has wave vector $\boldsymbol{k}_2 - \boldsymbol{k}_2 + \boldsymbol{k}_1$ or $\boldsymbol{k}_3 - \boldsymbol{k}_3' + \boldsymbol{k}_1$ responsible for the FWM signal in the four-level UMS, can be calculated by following the perturbation channels:

$$\text{(I)}\ \rho_{00}^{(0)} \xrightarrow{\varepsilon_1} \rho_{10}^{(1)} \xrightarrow{\varepsilon_2} \rho_{20}^{(2)} \xrightarrow{(\varepsilon_2')^*} \rho_{10}^{(3)},$$

$$\text{(II)}\ \rho_{00}^{(0)} \xrightarrow{\varepsilon_1} \rho_{10}^{(1)} \xrightarrow{\varepsilon_3} \rho_{30}^{(2)} \xrightarrow{(\varepsilon_3')^*} \rho_{10}^{(3)}.$$

While the fifth-order off-diagonal density matrix element $\rho_{10}^{(5)}$, which has wave vectors $\boldsymbol{k}_1 + 2\boldsymbol{k}_2 - 2\boldsymbol{k}_2', \boldsymbol{k}_1 + \boldsymbol{k}_2 - \boldsymbol{k}_2' + \boldsymbol{k}_3 - \boldsymbol{k}_3'$ and $\boldsymbol{k}_1 + 2\boldsymbol{k}_3 - 2\boldsymbol{k}_3'$ responsible for the SWM signal in the four-level UMS, can be calculated by following the perturbation channels:

$$\text{(III)}\rho_{00}^{(0)} \xrightarrow{\varepsilon_1} \rho_{10}^{(1)} \xrightarrow{\varepsilon_2} \rho_{20}^{(2)} \xrightarrow{(\varepsilon_2')^*} \rho_{10}^{(3)} \xrightarrow{\varepsilon_2} \rho_{20}^{(4)} \xrightarrow{(\varepsilon_2')^*} \rho_{10}^{(5)},$$

$$\text{(IV)}\rho_{00}^{(0)} \xrightarrow{\varepsilon_1} \rho_{10}^{(1)} \xrightarrow{\varepsilon_2} \rho_{20}^{(2)} \xrightarrow{(\varepsilon_2')^*} \rho_{10}^{(3)} \xrightarrow{\varepsilon_3} \rho_{30}^{(4)} \xrightarrow{(\varepsilon_3')^*} \rho_{10}^{(5)}.$$

$$\text{(V)}\rho_{00}^{(0)} \xrightarrow{\varepsilon_1} \rho_{10}^{(1)} \xrightarrow{\varepsilon_3} \rho_{30}^{(2)} \xrightarrow{(\varepsilon_3')^*} \rho_{10}^{(3)} \xrightarrow{\varepsilon_2} \rho_{20}^{(4)} \xrightarrow{(\varepsilon_2')^*} \rho_{10}^{(5)},$$

$$\text{(VI)}\rho_{00}^{(0)} \xrightarrow{\varepsilon_1} \rho_{10}^{(1)} \xrightarrow{\varepsilon_3} \rho_{30}^{(2)} \xrightarrow{(\varepsilon_3')^*} \rho_{10}^{(3)} \xrightarrow{\varepsilon_3} \rho_{30}^{(4)} \xrightarrow{(\varepsilon_3')^*} \rho_{10}^{(5)}.$$

Here, $\varepsilon_i, \boldsymbol{k}_i(\varepsilon_i', \boldsymbol{k}_i')$ are the constant field amplitude and the wave vector of the ω_i component in beam 2 (beam 3), repectively. While $\varepsilon_1, \boldsymbol{k}_1$ are the field amplitude and the wave vector of beam 1. Channels (I) and (II) correspond to two two-photon FWM processes. Channels (III)–(VI) correspond to four two-photon SWM processes. We are only interested in

the beat signal with modulation frequency $\omega_3-\omega_2$, so there are two beating approaches (III) (IV) (V) and (IV) (V) (VI).

The FWM and SWM signals are proportional to the average of the absolute square of $P^{(3)}$ and $P^{(5)}$ over the random variable of the stochastic process $\langle|P^{(3)}|^2\rangle$ and $\langle|P^{(5)}|^2\rangle$, which involve fourth-and eighth-order coherence functions of $u_i(t)$ in phase-conjugation geometry, respectively. And $u_i(t)$ is a dimensionless statistical factor that contains phase and amplitude fluctuations. While the FWM and SWM signal intensities in DeBeer's self-diffraction geometry are related to the sixth-and tenth-order coherence functions of the incident fields, respectively. We assume that beam 2 (beam 3) is multimode thermal source, $u_i(t)$ has Gaussian statistics.[11] If we are only interested in the τ-dependent part of the signal, the FWM and SWM signal intensities can be well approximated by the absolute squares of the stochastic average of the polarization $|\langle P^{(3)}\rangle|^2$ and $|\langle P^{(5)}\rangle|^2$, respectively, which involve 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 beating signal.[2,3,6,7] Furthermore, assuming that beam 2 (beam 3) has Lorentzian line shape, we have $< u_i(t_1)u_i^*(t_2) >= \exp(-\alpha_i|t_1-t_2|)$ $i=2,3$. Here $\alpha_i=\delta\omega_i/2$with $\delta\omega_i$ being the linewidth of the laser with frequency ω_i.

We first consider the case that beams 2 and 3 are narrow band so that $\alpha_2 << \Gamma_{20}$ and $\alpha_3 << \Gamma_{30}$. For simplicity, here we neglect the Doppler effect. Performing the tedious integration, the third-order beat signal intensity from channels (I) and (II) is

$$\begin{aligned} I^{(3)}(\tau) \propto & |B_1|^2\exp(-2\alpha_2|\tau|)+|\eta B_2|^2\exp(-2\alpha_3|\tau|) \\ & +\exp[-(\alpha_2+\alpha_3)|\tau|] \\ & \cdot\{\eta B_1^* B_2\exp[-\mathrm{i}\Delta\boldsymbol{k}\cdot\boldsymbol{r}-\mathrm{i}(\omega_3-\omega_2)\tau] \\ & +\eta^* B_1 B_2^*\exp[\mathrm{i}\Delta\boldsymbol{k}\cdot\boldsymbol{r}+\mathrm{i}(\omega_3-\omega_2)\tau]\}, \quad (1) \end{aligned}$$

where B_1 and B_2 are constants; $\Delta\boldsymbol{k}=(\boldsymbol{k}_2-\boldsymbol{k}_2')-(\boldsymbol{k}_3-\boldsymbol{k}_3')$; $\eta=\dfrac{\mu_3^2}{\mu_2^2}\left[\dfrac{\varepsilon_3(\varepsilon_3')^*}{\varepsilon_2(\varepsilon_2')^*}\right]$; τ is the time delay of beam 3 with respect to beam 2; μ_2,μ_3 is the dipole-moment matrix element between $|1\rangle$ and $|2\rangle$, $|1\rangle$ and $|3\rangle$; Γ_{20} and Γ_{30} are the transverse relaxation rates of the transitions from $|0\rangle$ to $|2\rangle$, $|0\rangle$ to $|3\rangle$.

When beams 2 and 3 are narrow band, the fifth-order beat signal intensity from channels (III) (IV) (V) is

$$\begin{aligned} I^{(5)}(\tau) \propto & |B_3|^2\exp(-4\alpha_2|\tau|)+|\eta B_4|^2\exp[-2(\alpha_2 \\ & +\alpha_3)|\tau|]+\exp[-(3\alpha_2+\alpha_3)|\tau|] \\ & \cdot\{\eta B_3^* B_4\exp[-\mathrm{i}\Delta\boldsymbol{k}\cdot\boldsymbol{r}-\mathrm{i}(\omega_3-\omega_2)\tau] \\ & +\eta^* B_3 B_4^*\exp[\mathrm{i}\Delta\boldsymbol{k}\cdot\boldsymbol{r}+\mathrm{i}(\omega_3-\omega_2)\tau]\}, (2) \end{aligned}$$

where B_3 and B_4 are constants. Equations (1) and (2) indicate that signal intensity oscillate not only temporally with period $\omega_3-\omega_2$, but also spatially with period $2\pi/\Delta k$ along the direction $\Delta\boldsymbol{k}$, which is almost perpendicular to the propagation direction of the beat signal. Here $\Delta k\approx 2\pi|\lambda_2-\lambda_3|\theta/(\lambda_2\lambda_3)$, θ is the angle between beams 2 and 3. Physically, the polarization-beat model assumes that both pump beams are plane waves. Therefore, two two-photon FWM signals, which propagate along $\boldsymbol{k}_{s_1}=\boldsymbol{k}_2-\boldsymbol{k}_2'+\boldsymbol{k}_1$ and $\boldsymbol{k}_{s2}=\boldsymbol{k}_3-\boldsymbol{k}_3'+\boldsymbol{k}_1$, respectively, are plane waves also. Since the two two-photon FWM propagate along slightly different directions, the interference between them leads to the spatial oscillation. While Eqs. (1) and (2) indicate that the beat signals modulate with dampling rates $\alpha_2+\alpha_3$ and $3\alpha_2+\alpha_3$ as τ is varied, respectively. In the cases 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, 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 $\Omega_3-\Omega_2$. In fact, 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.[2,12]

It is interesting to understand the underlying physics in UMS with incoherent lights. Much attention has been paid to the study of various ultrafast phenomena by using incoherent light sources recently.[13] We then consider the case that beams 2 and 3 are broadband so that $\alpha_2 >> \Gamma_{20}$ and $\alpha_3 >> \Gamma_{30}$. In this case, the beat signal rises to its maximum quickly and then decays with a time constant mainly determined by the transverse relaxation time of the system. Although the beat signal modulation is complicated in general, at the tail of the signal (i.e. $\tau >> \alpha_2^{-1}, \tau >> \alpha_3^{-1}$) the third-order beat signal intensity from channels (I) and (II) is

$$I^{(3)}(\tau) \propto |B_5|^2 \exp(-2\Gamma_{20}|\tau|) + |\eta B_6|^2 \exp(-2\Gamma_{30}|\tau|) + \exp[-(\Gamma_{20}+\Gamma_{30})|\tau|] \times \{\eta B_5^* B_6 \exp[-\mathrm{i}\Delta\boldsymbol{k}\cdot\boldsymbol{r} - \mathrm{i}(\Omega_3-\Omega_2)\tau] + \eta^* B_5 B_6^* \exp[\mathrm{i}\Delta\boldsymbol{k}\cdot\boldsymbol{r} + \mathrm{i}(\Omega_3-\Omega_2)\tau]\}, \quad (3)$$

where B_5 and B_6 are constants.

When beams 2 and 3 are broad band, at the tail of the signal (i.e. $\tau >> \alpha_2^{-1}, \tau >> \alpha_3^{-1}$) the fifth-order beat signal intensity from channels (III) (IV) (V) is

$$I^{(5)}(\tau) \propto |B_7|^2 \exp(-4\Gamma_{20}|\tau|) + |\eta B_8|^2 \exp(-2(\Gamma_{20}+\Gamma_{30})|\tau|) + \exp[-(3\Gamma_{20}+\Gamma_{30})|\tau|] \cdot \{\eta B_7^* B_8 \exp[-\mathrm{i}\Delta\boldsymbol{k}\cdot\boldsymbol{r} - \mathrm{i}(\Omega_3-\Omega_2)\tau] + \eta^* B_7 B_8^* \exp[\mathrm{i}\Delta\boldsymbol{k}\cdot\boldsymbol{r} + \mathrm{i}(\Omega_3-\Omega_2)\tau]\}, \quad (4)$$

where B_7 and B_8 are constants. Equations (3) and (4) indicate that the modulation frequency of the beat signal equals $\Omega_3 - \Omega_2$. The overall accuracy of using TPBFS and FPBFS with broadband lights to measure the energy-level difference between two excited states which are dipolar forbidden from the ground state is limited by the homogeneous linewidths.[1,6,7]

III. DISCUSSION AND CONCLUSION

However, for highly nonlinear media such as semiconductors, polymers and some special glasses, high-order nonlinearities may also contribute even at moderate intensity levels. In principle, the interference effect may be observed in a large variety of materials and the exploitation of this phenomenon to develop a new technique for nonlinear spectroscopy is also discussed.[8,9,14] The signal moduation depth is dependent on the ratio $\chi^{(5)}/\chi^{(3)}$ and can be adjusted by changing the lasers' relative intensities,[10] where $\chi^{(3)}$ and $\chi^{(5)}$ are third-and fifth-order susecptibilities, respectively. Based on Eqs. (1) and (2), we have calculated the TPBFS and FPBFS in sodium vapor, where the ground state $3S_{1/2}$, the intermediate state $3P_{1/2}$, and two excited states $6S_{1/2}$ and $5D_{3/2}$ formed a four-level system. In SI units, we inferred the ratio between third-and fifth-order beat modulation depths $\left(\frac{\chi^{(5)}}{\chi^{(3)}}\right)^2 \frac{[I(\omega_2)I(\omega_3)]^2}{I(\omega_2)I(\omega_3)}$=0.1, where $I(\omega_i)$ is the laser intensity of the ω_i component. Figure 3 presents the temporal behavior of normalized beat signal intensity versus relative time delay. The dashed curve and solid curve correspond to third-and fifth-order polarization beats, respectively. Parameters are α_2=3.6ps^{-1}, α_3=3.8ps^{-1}, λ_2=514.9nm, λ_3=497.9nm, Δk=0 η=B_1=B_2=1, B_3=B_4=0.32. It shows that as τ is varied, the beat signal intensity modulates sinusoidally with a period 50fs. The modulation frequency can be obtained more directly by making a Fourier transformation of the TPBFS and PBFS data. Then we obtain the modulation frequency 125ps^{-1}, corresponding to the beating between the resonant frequencies of the transitions from $3P_{1/2}$ to $6S_{1/2}$ to $5D_{3/2}$. Figure 4 shows the spatially behavior of normalized third-order polarization beat signal intensity versus transverse distance r. Parameters are $\alpha_2 = 3.6\mathrm{ps}^{-1}$, $\alpha_3 = 3.8\mathrm{ps}^{-1}$, $\tau = 1$ps, $\theta = 2.62 \times 10^{-2}$ rad, $\lambda_2 = 514.9$nm, $\lambda_3 = 497.9$nm, $\eta = B_1 = B_2 = 1$.

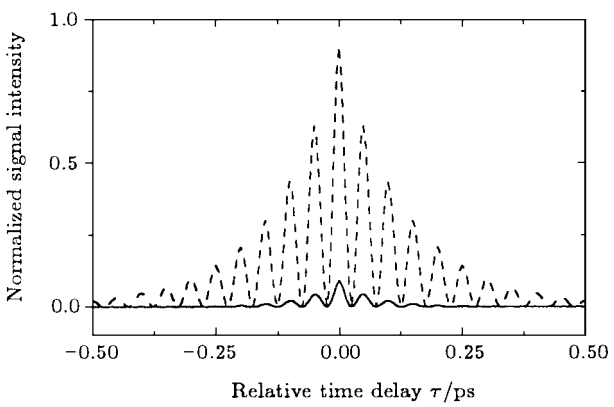


Fig.3. Normalized beat signal intensity versus relative time delay. The dashed curve and solid curve correspond to third-and fifth-order polarization beats, respectively. Parameters are α_2=3.6ps^{-1}, α_3=3.8ps^{-1}, λ_2=514.9nm, λ_3=497.9nm, η=B_1=B_2 = 1, Δk=0, B_3=B_4=0.32.

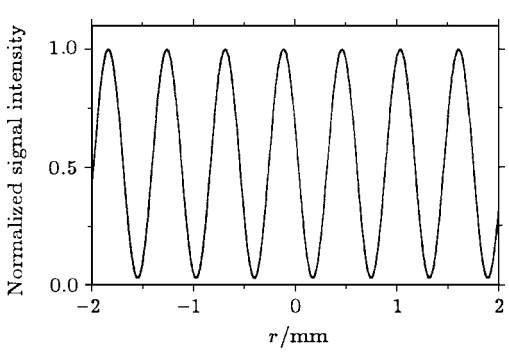


Fig.4. Normalized third-order polarization beat signal intensity versus transverse distance r. Parameters are α_2=3.6ps^{-1}, α_3=3.8ps^{-1}, τ=1ps, θ=2.62 × 10^{-2} rad, λ_2=514.9nm, λ_3=497.9nm, η=B_1=B_2=1.

In conclusion, we have employed a second-order coherence function theory to study the nonlinear effects of third-and fifth-order on polarization beats in

a four-level system. It is found that, the different temporal behavior of the beat signal in TPBFS and FPBFS depends on the stochastic properties of the lasers and transverse relaxation rate of the transition. We have considered the cases that pump beams have either narrow band or broadband linewidth and found that for both cases a Doppler-free precision in the measurement of the energy-level difference between two excited states which are dipolar forbidden from the ground state can be achieved. It is worth mentioning that the asymmetric behavior of the polarization beat signal, which is due to the dispersion of the optical components, does not affect the overall accuracy of using TPBFS and FPBFS to measure the energy-level difference.[2,12] Furthermore, TPBFS and FPBFS 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 $c/|\omega_3 - \omega_2|$.

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