Ultrafast modulation spectroscopy in a cascade three-level system

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Based on the polarization interference between one-photon and two-photon processes, we demonstrated an ultrafast modulation spectroscopy in a cascade three-level system (UMSCTS) in Na vapor, which showed a modulation of the four-wave mixing signal intensity with a 55-fs period, corresponding to the beating between the resonant frequencies of the transitions from $3S_{1/2}$ to $3P_{3/2}$ and from $3P_{3/2}$ to $4D_{3/2,5/2}$. We have also discussed the possibility of using UMSCTS to measure the energy levels of the excited states with Doppler-free precision.

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Quantum beat spectroscopy has become a powerful tool for precise measurements of energy-level splittings since the advent of laser techniques in the 1960s. It appears in the conventional time-resolved fluorescence [1] and in the time-resolved nonlinear laser spectroscopy [2], such as time-delayed four-wave mixing (FWM) or photon echo. A common requirement for quantum beat techniques is that the excitation laser pulses have a spectral width larger than the energy-level splittings so that energy sublevels can be excited simultaneously. As an alternative of quantum beat nonlinear laser spectroscopy, DeBeer *et al.* [3] demonstrated recently an ultrafast modulation spectroscopy (UMS) in which the energy-level splitting between states can be widely separated in frequency compared with the bandwidth of the excitation laser pulses. Fu *et al.* [4] 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 when pump beams had either narrow-band or broadband linewidth.

Up to now, quantum beat spectroscopy has been applied to the quasi-two-level system in which the excited and the ground states consist of sublevel structures. In this letter, we report an ultrafast modulation spectroscopy in a cascade three-level system (UMSCTS). We observed for the first time, to the best of our knowledge, 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 the excited states can be deduced. The present work does not attempt to explore the limit of the measurement accuracy, but rather demonstrates the promise of this new technique.

UMSCTS is a polarization beat phenomenon [3,5–7] originating from the interference between one-photon and two-photon processes. Let us consider a cascade three-level system [Fig. 1(a)] with a ground state $|0\rangle$, an intermediate state $|1\rangle$, and an excited state $|2\rangle$. States between $|0\rangle$ and $|1\rangle$ and between $|1\rangle$ and $|2\rangle$ are coupled by dipolar transition with resonant frequencies Ω_1 and Ω_2, respectively, while states between $|0\rangle$ and $|2\rangle$ are dipolar forbidden. We consider in this cascade three-level system a double-frequency FWM experiment in which beams 1 and 2 consist of two frequency components ω_1 and ω_2, while beam 3 has frequency ω_3 [Fig. 1(b)]. We assume that $\omega_1 \simeq \Omega_1$ ($\omega_3 \simeq \Omega_1$) and $\omega_2 \simeq \Omega_2$, therefore $\omega_1(\omega_3)$ and ω_2 will drive the transitions from $|0\rangle$ to $|1\rangle$ and from $|1\rangle$ to $|2\rangle$, respectively. There are two processes involved in this double-frequency FWM. First, the ω_1 frequency component of beams 1 and 2 induce population gratings of states $|0\rangle$ and $|1\rangle$, which are probed by beam 3 of frequency ω_3. This is one-photon resonant FWM and the signal (beam 4) has frequency ω_3. Second, beam 3 and the ω_2 frequency component of beam 1 induce a coherence between $|0\rangle$ and $|2\rangle$ by two-photon transition, which is then probed by the ω_2 frequency component of beam 2. This is a two-photon FWM with a resonant intermediate state and the frequency of the signal equals ω_3 again. We are interested in the dependence of the FWM signal in-

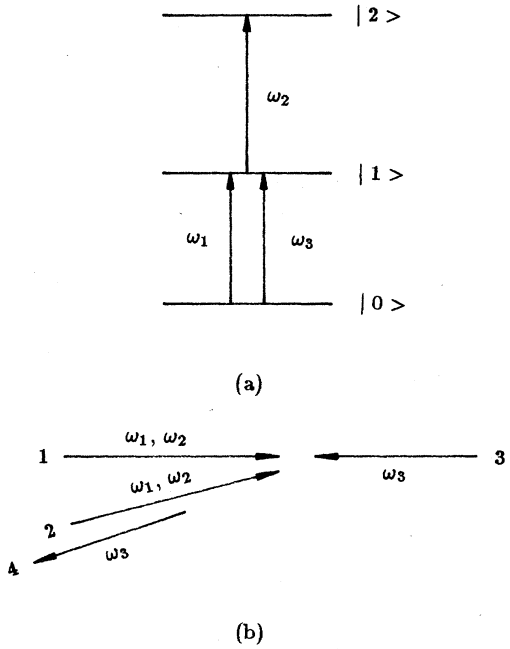

FIG. 1. (a) Three-level configuration to be treated in UMSCTS. (b) Schematic diagram of the geometry of UMSCTS.

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tensity as a function of the relative time delay between beams 1 and 2.

We assume that all incident beams are monochromatic lights. The complex electric fields of beam 1, E_{p1}, and beam 2, E_{p2}, can be written as $E_{p1}(\mathbf{r},t)=\varepsilon_1\exp[i(\mathbf{k}_1\cdot\mathbf{r}-\omega_1 t)]+\varepsilon_1\exp[i(\mathbf{k}_2\cdot\mathbf{r}-\omega_2 t)]$, $E_{p2}(\mathbf{r},t)=\varepsilon_1'\exp[i(\mathbf{k}_1'\cdot\mathbf{r}-\omega_1 t+\omega_{-1}\tau)]+\varepsilon_2'\exp[i(\mathbf{k}_2'\cdot\mathbf{r}-\omega_2 t+\omega_2\tau)]$. Here ε_i, $\mathbf{k}_i(\varepsilon_i',\mathbf{k}_i')$ are the constant field amplitude and the wave vector of the ω_i component in beam 1 (beam 2), respectively; τ is the relative time delay between beams 1 and 2. On the other hand, the complex electric field of beam 3 can be written as $E_3(\mathbf{r},t)=\varepsilon_3\exp[i(\mathbf{k}_3\cdot\mathbf{r}-\omega_3 t)]$. Here, ε_3 and $\mathbf{k}_3$ are the field amplitude and the wave vector of the field, respectively. The FWM signal is related to the third-order off-diagonal density-matrix element $\rho_{10}^{(3)}$, which has wave vector $\mathbf{k}_1-\mathbf{k}_1'+\mathbf{k}_3$ or $\mathbf{k}_2-\mathbf{k}_2'+\mathbf{k}_3$. It can be calculated by the following perturbation chains:

$$\text{(I)}\quad \rho_{00}^{(0)}\xrightarrow{\varepsilon_1}\rho_{10}^{(1)}\xrightarrow{(\varepsilon_1')^*}\rho_{00}^{(2)}\xrightarrow{\varepsilon_3}\rho_{10}^{(3)},$$

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

$$\text{(III)}\quad \rho_{00}^{(0)}\xrightarrow{\varepsilon_1}\rho_{10}^{(1)}\xrightarrow{(\varepsilon_1')^*}\rho_{11}^{(2)}\xrightarrow{\varepsilon_3}\rho_{10}^{(3)},$$

$$\text{(IV)}\quad \rho_{00}^{(0)}\xrightarrow{(\varepsilon_1')^*}(\rho_{10}^{(1)})^*\xrightarrow{\varepsilon_1}\rho_{11}^{(2)}\xrightarrow{\varepsilon_3}\rho_{10}^{(3)},$$

and

$$\text{(V)}\quad \rho_{00}^{(0)}\xrightarrow{\varepsilon_3}\rho_{10}^{(1)}\xrightarrow{\varepsilon_2}\rho_{20}^{(2)}\xrightarrow{(\varepsilon_2')^*}\rho_{10}^{(3)}\ .$$

Chains (I)–(IV) correspond to the one-photon resonant FWM, while chain (V) corresponds to the two-photon FWM. For simplicity, here we neglect the Doppler effect. The FWM signal intensity then becomes

$$I(\tau)\propto|B_1|^2+|\eta B_2|^2+\eta B_1^* B_2\exp[-i(\omega_2-\omega_1)\tau]+\eta^* B_1 B_2^*\exp[i(\omega_2-\omega_1)\tau], \quad (1)$$

with $B_1=2\Gamma_{10}(\Gamma_0+\Gamma_1)/\Gamma_0\Gamma_1(\Gamma_{10}+i\Delta_3)(\Gamma_{10}^2+\Delta_1^2)$, $B_2=1/(\Gamma_{10}+i\Delta_3)^2[\Gamma_{20}+i(\Delta_2+\Delta_3)]$, and $\eta\simeq(\mu_2/\mu_1)^2\times[\varepsilon_2(\varepsilon_2')^*/\varepsilon_1(\varepsilon_1')^*]$. Here, $\mu_1(\mu_2)$ is the dipole moment matrix element between $|0\rangle$ and $|1\rangle$ ($|1\rangle$ and $|2\rangle$); $\Delta_1=\Omega_1-\omega_1$, $\Delta_2=\Omega_2-\omega_2$, $\Delta_3=\Omega_1-\omega_3$; $\Gamma_0(\Gamma_1)$ is the longitudinal relaxation rate of the state $|0\rangle$ ($|1\rangle$); $\Gamma_{10}(\Gamma_{20})$ is the transverse relaxation rate of the coherence between states $|0\rangle$ and $|1\rangle$ ($|0\rangle$ and $|2\rangle$). Equation (1) indicates that the FWM signal modulates with a frequency $\omega_2-\omega_1$ as τ is varied. In the case that ω_1 and ω_2 are tuned to the resonant frequencies of the transitions from $|0\rangle$ to $|1\rangle$ and from $|1\rangle$ to $|2\rangle$, respectively, then the modulation frequency equals $\Omega_2-\Omega_1$. In other words, with UMSCTS we can obtain beating between the resonant frequencies of a cascade three-level system.

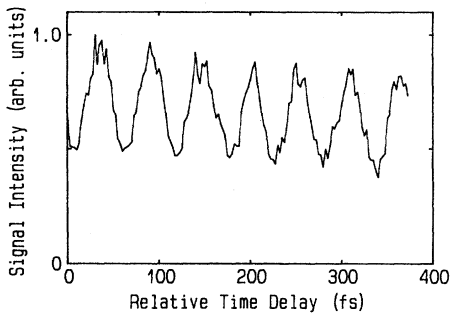

FIG. 2. Experimental result of the FWM signal intensity vs relative time delay.

We performed UMSCTS in sodium vapor, where the $3S$ ground state, $3P$ intermediate state, and $4D$ excited state formed a cascade three-level system. The Na vapor was contained in a stainless-steel heat-pipe oven with 0.5 torr argon as a buffer gas. The oven temperature was 210 °C. Two dye lasers (DL1 and DL2) pumped by the second harmonic of a Quanta-Ray YAG (yttrium aluminum garnet) laser, were used to generate frequencies at ω_1 and ω_2. DL1 and DL2 had linewidth 0.01 nm and pulse width 5 ns. The energies of DL1 and DL2 were about 3 and 1 mJ, respectively. DL1 was tuned to 589.0 nm, the wavelength of the $3S_{1/2}-3P_{3/2}$ transition, while DL2 was tuned to 568.8 nm, the wavelength of the $3P_{3/2}-4D_{3/2,5/2}$ transition. A beam splitter was used to combine the ω_1 and ω_2 components for beams 1 and 2, which intersected in the oven with a small angle (0.5°) between them. The relative time delay between beam 1 and beam 2 could be varied by an optical delay line controlled by a stepping motor. Beam 3, which propagated along the direction opposite that of beam 1, was derived from the DL1. All the incident beams were linearly polarized in the same direction. They were focused to spots with diameters of approximately 0.8 mm. The FWM signal, which had the same polarization as the incident beams, propagated along a direction almost opposite that of beam 2. It was detected by a photodiode and then fed into a signal averager for data averaging. A computer was used for data processing and for controlling the stepping motor to vary the relative time delay.

We first performed a degenerate FWM experiment with beams 1 and 2 consisting of the ω_1 frequency component. From the degenerate FWM spectrum we tuned ω_1 to the resonant frequency Ω_1. We then performed a nondegenerate FWM experiment in which beams 1 and 2 consisted of the ω_2 frequency component. We measured the nondegenerate FWM spectrum by scanning ω_2, which showed a resonant profile due to the two-photon transition. From this spectrum ω_2 was tuned to the resonant frequency Ω_2. After that, we performed the UMSCTS experiment by measuring the FWM signal intensity as a function of the relative time delay τ when beams 1 and 2 consisted of both frequencies ω_1 and ω_2. Figure 2 presents the result of a typical measurement. It shows that as τ is varied, the FWM signal intensity modulates sinusoidally with period 55 fs. The modulation frequency can be obtained more directly by making a Fourier transformation of the UMSCTS data. Figure 3 presents the Fourier spectrum of the data in which τ is varied for a range of 15 ps. From the Fourier spectrum we obtain the modula-

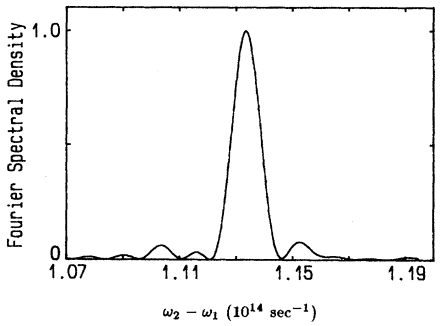

FIG. 3. Fourier spectrum of the UMSCTS data in which τ is varied for a range of 15 ps.

tion frequency $\omega_2-\omega_1=(1.134\pm0.005)\times10^{14}$ s^{-1}, corresponding to the beating between the resonant frequencies of the transitions from $3S_{1/2}$ to $3P_{3/2}$ and from $3P_{3/2}$ to $4D_{3/2,5/2}$.

As mentioned before, UMSCTS can be used to measure the energy separation between states $|1\rangle$ and $|2\rangle$ if the energy separation between $|0\rangle$ and $|1\rangle$ is well known. We first consider the case that the laser linewidths are much narrower than the homogeneous linewidths of the transitions. The measurement accuracy depends on the accuracy in measuring the FWM modulation frequency and the precision in tuning ω_1 and ω_2 to Ω_1 and Ω_2, respectively. The accuracy in the modulation frequency measurement can be improved by simply increasing the range of the time delay. However, due to the phase fluctuations of the laser fields, the amplitude of the signal modulation decays with a time constant determined by the laser coherence times as $|\tau|$ is increased [8]. As a result, the theoretical limit of the modulation frequency measurement is determined by the laser linewidths. For narrow-band laser sources the modulation frequency can be measured with high accuracy. In this case, the precision of using UMSCTS to measure $\Omega_2-\Omega_1$ is determined by how well ω_1 and ω_2 can be tuned to Ω_1 and Ω_2, respectively. Let beam 3 and the ω_1 component of the pump beams originate from the same laser source (i.e., $\omega_3=\omega_1$). Similar to the saturated absorption spectroscopy, one-photon resonant degenerate FWM can provide a Doppler-free spectrum with a peak located at $\Delta_1=0$ [9]. When ω_1 is set to the center of the Doppler profile, then only atoms whose velocities are $\mathbf{k}_1\cdot\mathbf{v}\simeq0$ interact with beam 3. This group of atoms will interact with beam 1 of frequency ω_2 and contribute to the two-photon nondegenerate FWM signal. Since only atoms in a specific velocity group contribute to the signal, the two-photon nondegenerate FWM spectrum is also Doppler-free. A similar situation in the two-photon absorption with a resonant intermediate state has been discussed by Bjorkholm and Liao [10]. Combining the capability of the high accuracy in measuring the FWM modulation frequency, a Doppler-free precision can be achieved in the measurement of $\Omega_2-\Omega_1$.

We now consider the case that beams 1 and 2 are broadband so that $\alpha_1\gg\Gamma_{10}$ and $\alpha_2\gg\Gamma_{20}$. Here $\alpha_i=\delta\omega_i/2$ with $\delta\omega_i$ the linewidth [full width at half maximum (FWHM)] of the laser with frequency ω_i. Based on the technique developed in Ref. [4] we study the temporal behavior of the FWM signal intensity in the extremely Doppler-broadened limit. It is found that the FWM signal rises to its maximum quickly and then decays with time constant mainly determined by the transverse relaxation times of the system. If we tune the frequency ω_3 of beam 3, which is assumed to be monochromatic, to the center of the Doppler profile by performing degenerate FWM or saturated absorption spectroscopy, then we have at the tail of the signal (i.e., $\tau\gg\alpha_1^{-1},\alpha_2^{-1}$)

$$
\begin{aligned}
I(\tau)\propto{}& C_1^2\left(\frac{1}{\Gamma_0}+\frac{1}{\Gamma_1}\right)^2\exp(-2\Gamma_{10}^a|\tau|)\\
&+|\eta|^2C_2^2\tau^2\exp(-2\Gamma_{20}^a|\tau|)\\
&+C_1C_2\tau\left(\frac{1}{\Gamma_0}+\frac{1}{\Gamma_1}\right)\exp[-(\Gamma_{10}^a+\Gamma_{20}^a)|\tau|]\\
&\times(\eta\exp[-i(\Omega_2-\Omega_1)\tau]+\eta^*\exp[i(\Omega_2-\Omega_1)\tau]).
\end{aligned}
\tag{2}
$$

Here, $C_1=\alpha_1/(\alpha_1^2+\Delta_1^2)$, $C_2=(\zeta_2-1)\alpha_2/(\alpha_2^2+\Delta_2^2)$, $\Gamma_{10}^a=(1+\zeta_1)\Gamma_{10}$, $\Gamma_{20}^a=\Gamma_{20}+(\zeta_2-1)\Gamma_{10}$, $\zeta_1=k_1/k_3$, and $\zeta_2=k_2/k_3$. In deriving Eq. (2) we assume $\zeta_2>1$. According to Eq. (2), the FWM signal exhibits a damping oscillation with frequency $\Omega_2-\Omega_1$ and a damping rate $\Gamma_{10}^a+\Gamma_{20}^a$. Therefore the modulation frequency, *which corresponds directly to the beating between the resonant frequencies of the cascade three-level system*, can be measured with an accuracy given by $\pi[\Gamma_{10}^a+\Gamma_{20}^a]$ approximately. In other words, UMSCTS with broadband lights is again a Doppler-free spectroscopy. The main difference between UMSCTS with narrowband lights and broadband lights is that for the former case the modulation frequency is $\omega_2-\omega_1$, while for the latter case the modulation frequency is $\Omega_2-\Omega_1$. Physically, as pointed out by Morita and Yajima [11], the time-delayed FWM with broadband light is related intrinsically to the optical coherent transient spectroscopy. In our case the one-photon and two-photon FWM in UMSCTS are related to the three-pulse stimulated photon echo and sum-frequency trilevel echo [12], respectively. As coherent transient effects there exists free evolution of the dipole moment and the two-photon coherence. Therefore, the modulation in the broadband-light UMSCTS reflects the characteristic of the system directly.

UMSCTS 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, UMSCTS is closely related to the Doppler-free saturated absorption spectroscopy and two-photon absorption spectroscopy in tuning ω_1 and ω_2 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 lasers do not need to be well calibrated.* In this sense UMSCTS is similar to the spectroscopies in the time domains. UMSCTS is related intrinsically to the optical coherent transient spectroscopy when broadband lights are used. The advantage of UMSCTS over other time-domain techniques is that *the temporal resolution is not limited by the laser pulse width.* For example, in this experiment we observed a beat note of 55-fs period even when the lasers had a pulse width of 5 ns. Finally, since the pioneering work of Morita and Yajima [11] time-delayed FWM with broadband lights has attracted much

attention. Up to now, this technique has been concentrated on studying ultrafast relaxation processes in the time domain with a resolution that is better than the laser pulse width [13]. In this letter, we point out that through interference between two FWM processes, the time-delayed FWM with broadband lights can even be used as a Doppler-free high-resolution spectroscopy. It is worth mentioning that, from a practical experimental consideration, since ω_1 and ω_2 frequency components of beams 1 and 2 propagate along the same optical path and use the same optical components, UMSCTS 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 $c/|\omega_2-\omega_1|$.

In conclusion, we demonstrated UMSCTS in Na vapor, which showed a modulation of the FWM signal intensity with a period of 55 fs, corresponding to the beating between the resonant frequencies of the transitions from $3S_{1/2}$ to $3P_{3/2}$ and from $3P_{3/2}$ to $4D_{3/2,5/2}$. We discussed the possibility of using UMSCTS to measure the energy levels of the excited states with Doppler-free precision.

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