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Field-Correlation Effects on Rayleigh-Enhanced Nondegenerate Four-Wave Mixing *

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We study Rayleigh-enhanced nondegenerate four-wave mixing (NFWM) with time-delayed, correlated fluctuating fields. The importance of the field correlation is revealed in the Rayleigh-enhanced NFWM spectrum when the time delay is varied. The Rayleigh-enhanced NFWM is employed to study the ultrafast processes in the frequency domain. A relaxation time as short as 220 fs was deduced in the Rayleigh-enhanced NFWM experiments in carbon disulphide.

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Lasers are inherently noisy devices, in which both the phase and amplitude of the field can fluctuate. There are many different statistical models to describe laser fields. However, since many models of fluctuating laser fields have identical second-order field correlations, differences among them will become important only if the observable depends on higher-order field correlations. Various nonlinear optical interaction, such as resonance fluorescence, four-wave mixing (FWM) and two-photon absorption, have been studied.[1−3] Recently, there has been considerable interest in the study of atoms interacting with time-delayed, correlated fluctuating fields. Optical transients induced by time-delayed correlated pulses have been proven to be a powerful technique in obtaining subpicosecond time resolution.[4−7] In this letter, we will study Rayleigh-enhanced nondegenerate four-wave mixing (NFWM) with time-delayed, correlated fluctuating fields.

Rayleigh-enhanced NFWM is a third-order nonlinear phenomenon with three incident beams involved. Beams 1 and 2 have the same frequency ω_1 and a small angle θ exists between them. Beam 3, with frequency ω_3, propagates along the opposite direction of beam 1. The NFWM signal has frequency ω_3 and propagates along the opposite direction of beam 2 approximately. The complex incident laser fields can be written as $E_i(\boldsymbol{r},t) = A_i(\boldsymbol{r},t)\exp(-\mathrm{i}\omega_i t) = \varepsilon_i u_i(t)\exp[\mathrm{i}(\boldsymbol{k}_i\cdot\boldsymbol{r} - \omega_i t)], (i = 1,2,3)$, where ε_i and $\boldsymbol{k}_i$ are the constant field magnitude and the wave vector of the ith beam, respectively, and u_i(t) is a dimensionless statistical factor that contains the phase and the amplitude fluctuations. We assume that beams 1 and 2 come from a single laser source, then $u_1(t) = u(t)$, $u_2(t) = u(t-\tau)$. Here τ is the time delay of beam 2 with respect to beam 1. The nonlinear polarization responsible for the Rayleigh-enhanced NFWM is $P(\boldsymbol{r},t) = P_1(\boldsymbol{r},t) + P_3(\boldsymbol{r},t)$ with $P_1(\boldsymbol{r},t) = Q_1(\boldsymbol{r},t)E_3(\boldsymbol{r},t)$ and $P_3(\boldsymbol{r},t) = Q_3(\boldsymbol{r},t)E_1(\boldsymbol{r},t)$. Here $Q_1(\boldsymbol{r},t)$ and $Q_3(\boldsymbol{r},t)$ are the order parameters of the gratings induced by beams 1 and 2, beams 2 and 3, respectively, which satisfy

$$\begin{aligned}\frac{\partial Q_1}{\partial t} + \gamma_1 Q_1 &= \gamma_1\chi_1 A_1 A_2^* \exp[\mathrm{i}(\boldsymbol{k}_1 - \boldsymbol{k}_2)\cdot\boldsymbol{r}],\\ \frac{\partial Q_3}{\partial t} + \gamma_3 Q_3 &= \gamma_3\chi_3 A_3 A_2^* \exp[\mathrm{i}(\boldsymbol{k}_3 - \boldsymbol{k}_2)\cdot\boldsymbol{r} - \Delta t].\end{aligned} \tag{1}$$

Here $\Delta = \omega_3 - \omega_1$, and χ_1 (χ_3) and γ_1 (γ_3) are the nonlinear susceptibility and relaxation rate of the grating induced by beam 1 (beam 3) and beam 2, respectively. Equation (1) can be solved formally, from which we can obtain

$$\begin{aligned}P_1(\boldsymbol{r},t) = S(\boldsymbol{r},t)[&\chi_1\gamma_1 A_3(t)\int_0^\infty \mathrm{d}t' A_1(t-t')\\ &\cdot A_2^*(t-t')\exp(-\gamma_1 t')],\\ P_3(\boldsymbol{r},t) = S(\boldsymbol{r},t)\{&\chi_3\gamma_3 A_1(t)\int_0^\infty \mathrm{d}t' A_2^*(t-t')\\ &\cdot A_3(t-t')\exp[-(\gamma_3 - \mathrm{i}\Delta)t']\}.\end{aligned} \tag{2}$$

Here $S(\boldsymbol{r},t) = \exp\{\mathrm{i}[(\boldsymbol{k}_1 - \boldsymbol{k}_2 + \boldsymbol{k}_3)\cdot\boldsymbol{r} - \omega_3 t]\}$. The FWM signal intensity is proportional to the average of the absolute square of the polarization over the random variable of the stochastic process, i.e. $I \propto \langle|P|^2\rangle$, which involves the second-order coherence function of $u_3(t)$ and fourth-order coherence function of $u(t)$. We assume that the pump laser is a multimode thermal source. In this case, $u(t)$ has Gaussian statistics with its fourth-order coherence function satisfying

$$\begin{aligned}\langle u(t_1)u(t_2)u^*(t_3)u^*(t_4)\rangle = &\langle u(t_1)u^*(t_3)\rangle\langle u(t_2)u^*(t_4)\rangle\\ &+ \langle u(t_1)u^*(t_4)\rangle\langle u(t_2)u^*(t_3)\rangle.\end{aligned}$$

If the laser sources have Lorentzian lineshape, we have the second-order coherence function

$$\begin{aligned}\langle u(t)u^*(t-\tau)\rangle &= \exp(-\alpha|\tau|),\\ \langle u_3(t)u_3^*(t-\tau)\rangle &= \exp(-\alpha_3|\tau|).\end{aligned}$$

Here $\alpha = \delta\omega_1/2$, $\alpha_3 = \delta\omega_3/2$ with $\delta\omega_1$ and $\delta\omega_3$ being the laser linewidths (full width at half maximum

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(FWHM)) of beams 1 and 3. Following the process given in Ref. [8], an analytic closed form for the FWM signal intensity $I(\Delta, \tau)$ can be obtained after tedious calculation.

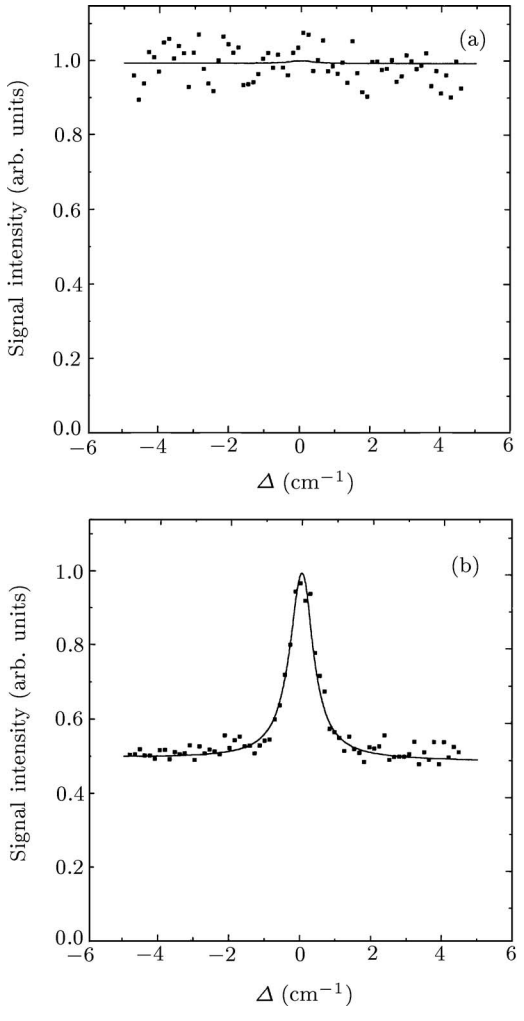


Fig. 1. Rayleigh-enhanced NFWM spectra in Oxazine dye when (a) $\tau = 0$ ps and (b) $\tau = 2800$ ps. The solid curves are the theoretical curves.

Two sets of experiments were performed to study the effects of field correlation. The second harmonic of a Quanta-Ray YAG laser was used to pump two dye lasers. The first dye laser (DL1) had wavelength 585 nm, linewidth 0.007 nm and pulsewidth 5 ns. It was split into beams 1 and 2 after passing through a beam splitter, and the two beams intersected in the sample with a small angle (1.3°) between them. Beam 3, originating from the second dye laser (DL2) with linewidth 0.007 nm, propagated along the direction opposite to that of beam 1. The wavelength of beam 3 could be scanned by a computer-controlled stepping motor. All the incident beams were focused to spots with diameters of approximately 0.5 mm. The NFWM signal, which propagated along a direction almost opposite to that of beam 2, 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 wavelength of beam 3.

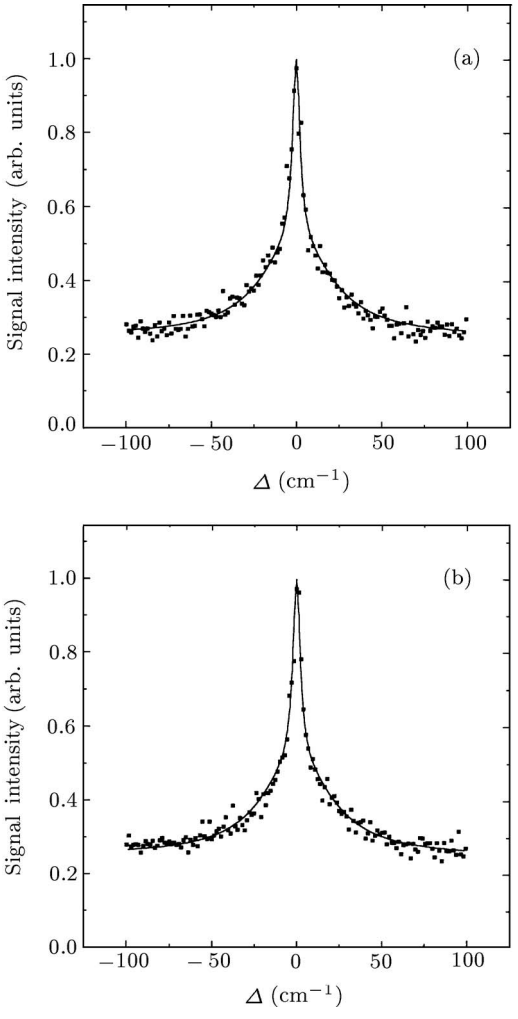


Fig. 2. Rayleigh-enhanced NFWM spectra in CS_2 when (a) $\tau = 0$ ps and (b) $\tau = 2800$ ps. The solid curves are the theoretical curves.

We first study Rayleigh-enhanced NFWM mediated by thermal effects. These experiments are in the broadband limit. Our sample was Oxazine dye dissolved in ethanol (2×10^{-4} mol). All the laser beams were vertically polarized. We kept the energy of DL1 and DL2 low enough (50 μJ) so that the induced thermal gratings were not saturated. Figures 1(a) and 1(b) present the Rayleigh-enhanced NFWM spectra when $\tau = 0$ and 2800 ps, respectively. The Rayleigh-enhanced NFWM spectrum exhibits a large nonresonant background which obscures the resonant signal almost completely when beams 1 and 2 are fully correlated (Fig. 1(a)). In contrast, the intensity of the resonant signal and the nonresonant background are about the same when beams 1 and 2 are uncorrelated (Fig. 1(b)). We note that the main source of the noise comes from the fluctuation of the laser fields. The solid curves in Fig. 1 are the theoretical curves. The thermal grating has a long relaxation time (in the microsecond region), which is larger than the

laser pulsewidth. Therefore, due to the finite interaction time between the laser and the material, the role of the relaxation time should be replaced by the laser pulsewidth. We have $\gamma_1 = \gamma_3 = \tau_p^{-1}$, where $\tau_p = 5 \times 10^{-9}$ s is the laser pulsewidth. On the other hand, we choose $\alpha = \alpha_3 = 3.8 \times 10^{10}\,\mathrm{s}^{-1}$ for the fitting, which are consistent with the independent measurements of the laser linewidths.

The sample used in our second experiments was CS_2 which was contained in a cell with a thickness of 2 mm. The output energies of the first and second lasers were 1.5 mJ and 1 mJ, respectively. In this experiment the incident fields had the following polarization configuration, $\boldsymbol{E}^{(1)} || \boldsymbol{E}^{(3)} || \boldsymbol{x}$ and $\boldsymbol{E}^{(2)} || \boldsymbol{y}$, and the signal was polarized along $\boldsymbol{y}$. As is well known, the optical Kerr effect for the liquid CS_2 has at least two components, i.e. a relatively long "Debye" component and a shorter "interaction-induced" component, which have relaxation times of about 2 ps and 300 fs.[9] Since the laser coherent time was about 50 ps, these experiments were in the narrow-bandwidth limit. Figures 2(a) and 2(b) present the Rayleigh-enhanced NFWM spectra when $\tau = 0$ and 2800 ps, respectively. It shows that the field correlation has little effect on the Rayleigh-enhanced NFWM spectrum. The resonant signal is larger than the nonresonant background by a factor of about three. To fit our experimental data, we extend our theory to include two relaxation components with relaxation times γ_f and γ_s. The ratio between these two components is η, so that for the monochromatic incident lights, the nonlinear polarization $P \propto \eta[1+\gamma_s/(\gamma_s - \mathrm{i}\Delta)] + (1-\eta)[1+\gamma_f/(\gamma_f - \mathrm{i}\Delta)]$. The solid curves are the theoretical curves with fitting parameters, $\eta = 0.3$, $\gamma_s = 3.0 \times 10^{11}\,\mathrm{s}^{-1}$ and $\gamma_f = 2.3 \times 10^{12}\,\mathrm{s}^{-1}$, which correspond to relaxation times of 1.7 ps and 220 fs for the molecular reorientation and interaction-induced effect, respectively. Other parameters are $\alpha = \alpha_3 = 3.8 \times 10^{10}\,\mathrm{s}^{-1}$.

The τ-dependence of the Rayleigh-enhanced NFWM spectra can be explained if we consider the phase fluctuation of the incident fields. The resonant signal and the nonresonant background originate from the order parameters $Q_3(\boldsymbol{r}, t)$ and $Q_1(\boldsymbol{r}, t)$, which are induced by beams 2 and 3 and beams 1 and 2, respectively. According to Eq. (2), integration effects are involved in the establishment of the order parameters of the gratings. We first consider the broadband case (i.e. $\gamma_1, \gamma_3 \ll \alpha, \alpha_3$). The effect of integration in the establishment of order parameters $Q_1(\boldsymbol{r}, t)$ and $Q_3(\boldsymbol{r}, t)$, which last approximately for a duration of the relaxation time γ_1^{-1} and γ_3^{-1}, respectively, is to wash out the gratings. At zero time delay, no washout takes place in the establishment of $Q_1(\boldsymbol{r}, t)$ because the phase factor ϕ_1 of $A_1(t-t')A_2^*(t-t')$ is stationary. On the other hand, the phase factor ϕ_3 of $A_3(t-t')A_2^*(t-t')$ is a random variable which fluctuates with a characteristic timescale $(\alpha + \alpha_3)^{-1}$. Due to the integration effect, the fast random fluctuation of ϕ_3 leads to the reduction of the amplitude of Q_3. Therefore, the Rayleigh-enhanced NFWM spectrum is dominated by a large nonresonant background when $\tau = 0$. The Rayleigh-enhanced NFWM spectrum in the limit of $\alpha|\tau| \gg 1$ is quite different. Similar to Q_3, Q_1 is now induced by mutually incoherent fields. If $\gamma_3 = \gamma_1$ and $\alpha_3 = \alpha$, then the influence of the integration effect on Q_1 and Q_3 are equal. In the case of $\chi_1 = \chi_3$ and $\Delta = 0$, the signals from Q_1 and Q_3 will be equal. Furthermore, the relative phase between $P_1(\boldsymbol{r}, t)$ and $P_3(\boldsymbol{r}, t)$ is a stochastic variable. Since there is no interference between them, we have $B = 1$. We now consider the case that beams 1 and 2 have narrow bandwidth. In this case, the material gratings have very short relaxation times, therefore, they can respond to the phase fluctuations of the fields almost immediately. In consequence, the field correlation has little influence on the Rayleigh-enhanced NFWM spectrum. More specifically, $A_1(t-t')A_2^*(t-t')$ and $A_2^*(t-t')A_3(t-t')$ in Eq. (2) are slowly varying functions in comparison with $\exp(-\gamma_1 t')$ and $\exp(-\gamma_3 t')$ which have peaks at $t' = 0$, therefore they can be approximated as $A_1(t)A_2^*(t)$ and $A_2^*(t)A_3(t)$, respectively. We have

$$P(\boldsymbol{r}, t) \propto A_1(t)A_2^*(t)A_3(t) \int_0^\infty \mathrm{d}t' \{\chi_1\gamma_1 \exp(-\gamma_1 t') + \chi_3\gamma_3 \exp[-(\gamma_3 - \mathrm{i}\Delta)t']\}.$$

The above equation indicates that the Rayleigh-enhanced NFWM spectrum is independent of τ. Although the phases of P_1 and P_3 fluctuate randomly, the relative phase between them is fixed. In the case of $\chi_1 = \chi_3$ and $\gamma_3 = \gamma_1$ we have $B = 3$, instead of $B = 1$ due to the interference between P_1 and P_3.

In conclusion, we have studied the field-correlation effects on Rayleigh-enhanced NFWM. Through measuring the FWM spectrum as a function of time-delayed, correlated fluctuating fields, the importance of the fourth-order field correlation has been revealed. We have also demonstrated that Rayleigh-enhanced NFWM is a powerful technique for measuring the ultrafast relaxation time in the frequency domain.

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