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Optical-density effect in heterodyne-detected accumulated photon echo

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The effect of optical density on the heterodyne-detected accumulated photon echo is investigated both experimentally and theoretically. The cause of the distortion of the echo temporal behavior observed in the $^4G_{5/2}$ manifold of Nd:YAG (where YAG is yttrium aluminum garnet) is attributed to the optical-density effect, which is closely related to the change of the field correlation function as the excitation pulse propagates through the sample.

Since the work of Hesselink and Wiersma,[1] the heterodyne-detected accumulated photon echo has become a powerful tool for measuring the phase relaxation time in the condensed matter. The most interesting feature of the accumulated photon echo has recently been revealed by Asaka, Nakatsuka, Fujiwara, and Matsuoka.[2] They have clarified that the signal strength of the accumulated photon echo can be formulated in terms of the field-correlation function of the excitation pulse. This approach leads to another practically important property of this method, that is, the heterodyne-detected signal gives a decay constant of $T_2/2$ regardless of the amount of inhomogeneous broadening. However, the previous investigation did not take account of the optical-density effect which means in the present case that the spectral shape, or equivalently, the field-correlation function of the excitation pulse, varies as the pulse propagates through the sample. We have found that in such instances the temporal behavior of the echo is distorted significantly because of the variation of the correlation function. This distortion becomes serious in an optically dense sample when the bandwidth of the excitation pulse largely exceeds the absorption spectral width. In particular, in a sample which exhibits a small inhomogeneous broadening compared to the homogeneous width, the signal decay constant does not yield the intrinsic T_2 time. In the opposite case with a large inhomogeneous broadening, however, T_2 is determined correctly from the exponential decay at longer delay times, although the distortion is serious at shorter delay times. Furthermore, we have found that this optical-density (OD) effect is identified with a negative signal which appears at the leading edge of the heterodyne-detected echo signal.

The OD effect[3,4] which was previously investigated in photon echo experiments is concerned with the change of the pulse area as a function of the distance the pulse traveled through the sample. Atoms at different depths in the sample experience different excitation amplitudes and shapes. This can profoundly affect the results of the pulsed optical coherence experiments. Such an OD effect can be avoided by lowering the excitation intensity as well as the optical absorbance. However, the present OD effect still affects the temporal behavior of the echo in the small-excitation-intensity limit since this effect originates from the change of the correlation function of the excitation pulse which always occurs so long as the laser bandwidth exceeds the absorption spectral width. Therefore, there is no way to avoid this OD effect except by lowering the optical absorbance of the sample. In this Rapid Communication, we report for the first time the results of both experiments and theoretical analyses on the OD effect in the heterodyne-detected accumulated photon echo.

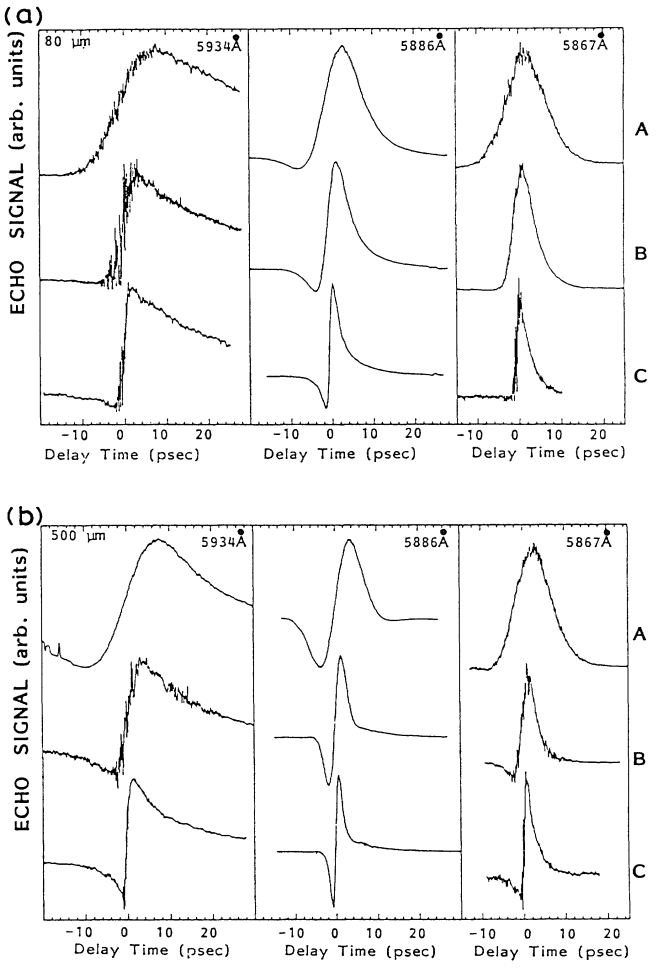

FIG. 1. Accumulated photon echos observed in three Stark levels of Nd-YAG at 10 K. The sample thickness is (a) 80 μm and (b) 500 μm. The dye laser bandwidth is 2.5 cm^{-1} in *A*, 7 cm^{-1} in *B*, and 25 cm^{-1} in *C*.

The OD effect on the accumulated photon echo was investigated for the $^4G_{5/2}$ manifold of Nd:YAG (where YAG is yttrium aluminum garnet). This manifold splits into three sharp lines in Nd:YAG.[5] The echo experiments were performed at liquid-helium temperature by means of a high-frequency-modulated heterodyne method using a synchronously pumped Rhodamine-6G dye laser. When the modulation frequency is raised over the decay rate of the bottleneck state $^4F_{3/2}$, the signal strength of the accumulated photon echo decreases to half its value at the low-frequency modulation.[6] However, it is advantageous to use the high-frequency-modulation method since the dye-laser jet-stream noise is significantly reduced as the frequency is raised to around 10 MHz.[7] An electro-optical or acousto-optical modulator was employed for the intensity modulation of the pump beam. Orthogonal linear polarization was used for the pump and probe beams so as to eliminate the thermal grating effect. The samples of Nd:YAG were 80- and 500-μm-thick pieces. From a transmission measurement on the 80-μm-thick sample at 10 K, the optical density and the spectral full width at half maximum (FWHM) of the three Stark lines were determined to be 0.2, 1.1, and 0.1 for the optical density, and 0.67, 1.7, and 2.5 cm^{-1} for the FWHM, corresponding to the 5934-, 5886-, and 5867-Å lines.

The accumulated photon echoes observed in the three Stark levels of Nd:YAG are shown in Figs. 1(a) and 1(b), corresponding to sample thicknesses of 80 and 500 μm, respectively. The dye-laser bandwidth was set to 2.5 cm^{-1} for signal A of Fig. 1, 7 cm^{-1} for signal B, and 25 cm^{-1} for signal C by changing the number of birefringent filters. The most prominent feature observed in the photon echo of Nd:YAG is that a negative signal or a dispersion-shaped signal appears near the origin of the delay time. The relative strength of the negative signal is enhanced as the optical density of the sample and the laser bandwidth are increased. In particular, in the 5886-Å line which exhibits the highest absorbance among the three lines, the dispersion-shaped signal predominates, masking the exponential decay. Although the logarithmic plot of this signal was not a straight line near the time origin, T_2 of this transition was determined to be 14 psec at 10 K from the exponential decay at longer delay times. T_2's for the other two lines of 5934 and 5867 Å were also determined to be 65 and 5 psec, respectively. As the laser bandwidth is increased over the absorption spectral width, the signal strength generally decreases since a small fraction of the spectrum within the laser bandwidth participates in the echo generation. However, it is advantageous to use an excitation pulse with a broader bandwidth in order to determine T_2 correctly. This is because, as will be discussed later, the OD affects the temporal behavior of the echo within the delay time corresponding to the inverse of the absorption spectral width.

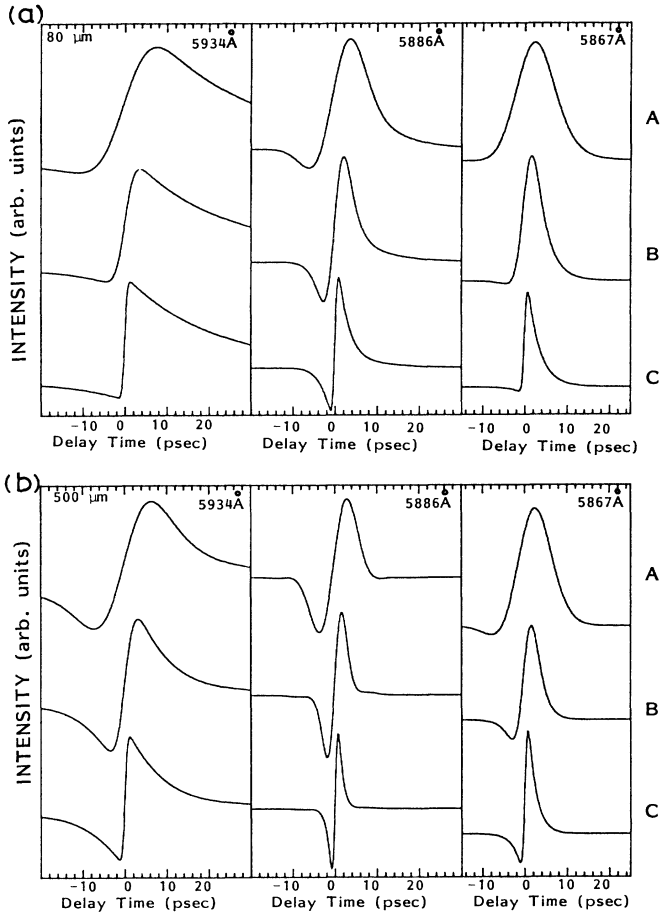

FIG. 2. Numerical simulations for the photon echos in three Stark levels of Nd:YAG. Each simulation corresponds to the echo signal in Fig. 1. Equation (7) was calculated using the spectral parameters of the three absorption lines.

We try to interpret theoretically the observed temporal behavior of the accumulated photon echo. The signal strength in pump-probe experiments is proportional to the product of the pump and probe intensities, in other words, proportional to the product of four optical fields. We designate the relevant electric fields as E_1 and E_3 for the pump beam and as E_2 and E_4 for the probe beam, where E_2 and E_4 are, respectively, the optically delayed replicas of E_1 and E_3. In the present calculation for the perturbation expansion of the density matrix, the fields E_1, E_2, and E_3 are associated with the third-order nonlinear polarization, and field E_4 corresponds to the probe field for heterodyne detection. From the third-order density-matrix expansion of the two-level system, the third-order nonlinear polarization is obtained as

$$P(t,z)=i\int_0^\infty d\tau\int_0^\infty d\tau'\int_0^\infty d\tau''\exp[-\tau'/T_1-(\tau+\tau'')/T_2]$$
$$\times\{E_3(t-\tau-t_{3,z})E_2(t-\tau-\tau'-t_{2,z})E_1^*(t-\tau-\tau'-\tau''-t_{1,z})$$
$$\times\exp[-(\tau-\tau'')^2\delta\omega^2/4-i\Omega(\tau-\tau'')]+E_3(t-\tau-t_{3,z})E_1^*(t-\tau-\tau'-t_{1,z})$$
$$\times E_2(t-\tau-\tau'-\tau''-t_{2,z})\exp[-(\tau+\tau'')^2\delta\omega^2/4-i\Omega(\tau+\tau'')]\}\ , \qquad (1)$$

where it has been assumed that the two-level system is characterized by the longitudinal and the transverse relaxation times T_1 and T_2, and the inhomogeneously broadened Gaussian line shape with center frequency Ω and the spectral width $\delta\omega$. If we assume a sufficiently long T_1, the write-in process due to E_1,E_2 pulses and the read-out process due to E_3,E_4 pulses can be temporally distinguished. This means that the write-in process is accumulated by the sequential excitation of picosecond pulses. Hence, we can, without losing generality, assume $t_4,t_3 \gg t_2,t_1$ where t_i is the arrival time of an E_i pulse. On these assumptions, the integral over τ' can be expressed as the field correlation function.

$$\int_0^\infty d\tau' E_2(t-\tau-\tau'-t_{2,z})E_1^*(t-\tau-\tau'-\tau''-t_{1,z}) = G^*(\tau''-t_{21,z}) \ ,$$
$$\int_0^\infty d\tau' E_1^*(t-\tau-\tau'-t_{1,z})E_2(t-\tau-\tau'-\tau''-t_{2,z}) = G^*(-\tau''-t_{21,z}) \ , \tag{2}$$

where $t_{21}=t_2-t_1$ is the delay time between the pump and probe beams. From the Wiener-Khintchine relation, the correlation function G^* can be expressed as

$$G^*(\tau,z) = \int I(\omega,z)\exp[-i(\omega+\omega_l)\tau]d\omega \ , \tag{3}$$

where ω_l is the center frequency of the laser spectrum and $I(\omega,z)$ is the power spectrum as a function of distance into the sample. By virtue of Eq. (2), Eq. (1) can be simplified as

$$P(t,z) = i\int_0^\infty d\tau \int_{-\infty}^\infty d\tau'' \exp[-(\tau+|\tau''|)/T_2]E_3(t-\tau-t_{3,z})G^*(\tau''-t_{21,z})\exp[-(\tau-\tau'')^2\delta\omega^2/4 - i\Omega(\tau-\tau'')] \ . \tag{4}$$

The variation of the field spectrum $E_i(\omega,z)$ as a function of distance is obtained from linearized Maxwell equations.

$$\frac{dE_i(\omega,z)}{dz} = -\tfrac{1}{2}\gamma(\omega)E_i(\omega,z), \text{ with } i=1,2,3 \ , \tag{5a}$$

$$\frac{dE_4(\omega,z)}{dz} = -\tfrac{1}{2}\gamma(\omega)E_4(\omega,z) - iP(\omega,z)\exp[-i(\omega+\omega_l)t_4] \ , \tag{5b}$$

where $E_i(\omega,z)$, $P(\omega,z)$, and the absorption spectrum $\gamma(\omega)$ are defined as

$$E_i(t,z) = \int d\omega E_i(\omega,z)\exp[-(\omega+\omega_l)t] \ ,$$
$$P(t,z) = \int d\omega P(\omega,z)\exp[-(\omega+\omega_l)t] \ , \tag{6}$$
$$\gamma(\omega) = \int dx \frac{\alpha}{(1/T_2)^2+(\Delta\omega-\omega-x)^2}\exp[-x^2/\delta\omega^2] \ ,$$

where $\Delta\omega$ denotes the frequency detuning and α is a constant including the transition dipole moment. In Eq. (5a), the backreaction of the induced nonlinear polarization on the fields E_i with $i=1,2,3$ is neglected due to the assumption of the sufficiently weak fields. Considering that the photodetected signal is expressed as $\int_{-\infty}^\infty dt\,|E_4(t-t_4,\,l)|^2$ or equivalently $\int_{-\infty}^\infty d\omega\,|E_4(\omega,l)|^2$ where l is the sample thickness, and also that the phase-sensitive detector picks up the signal which is linear in $P(\omega,l)$, we get the expression for the signal strength of the heterodyne-detected accumulated photon echo.

$$S = \mathrm{Re}\int_0^\infty d\tau \int_{-\infty}^\infty d\tau'' G_R(\tau-t_{21})G_W^*(\tau''-t_{21})\exp[-(\tau+|\tau''|)/T_2 - \delta\omega^2(\tau-\tau'')^2/4 - i\Delta\omega(\tau-\tau'')] \ , \tag{7}$$

with

$$G_R(\tau) = \int d\omega I(\omega)e^{-\gamma(\omega)l}e^{i\omega\tau} \ , \tag{8}$$

$$G_W(\tau) = \int d\omega I(\omega)\frac{1-e^{-\gamma(\omega)l}}{\gamma(\omega)}e^{i\omega\tau} \ , \tag{9}$$

where $I(\omega)$ is the power spectrum of the incoming excitation pulse. As was previously pointed out, the signal strength of the accumulated photon echo is formulated in terms of the field-correlation function of the excitation pulse. In the present heterodyne scheme, there are two modified correlation functions of G_W and G_R. G_W denotes the effective correlation function of the write-in process which induces the population change of the ground state. On the other hand, G_R denotes the effective correlation function of the read-out process through the heterodyne detection. It is evident that the correlation functions defined here are reduced to the usual Wiener-Khintchine relation provided the optical density of the sample is negligibly small.

We numerically evaluated Eq. (7) using the spectral parameters of the three absorption lines, all of which have been determined experimentally. As shown in Figs. 2(a) and 2(b), the numerical simulations corresponding to Fig. 1 reproduce the observed temporal behavior quite well with no adjustable parameters. This good agreement supports the validity of Eq. (7) and also confirms that the OD effect due to the change of the correlation function affects significantly the temporal behavior of the accumulated photon echo. By virtue of Eq. (7), we have further investigated the variation of the echo temporal behavior when several parameters including the frequency detuning, the laser bandwidth, the optical density, and the homogeneous

and inhomogeneous widths are varied. Here we present only the results concerned with the influence of the amount of optical density on the echo temporal behavior. We considered two kinds of systems which have the same homogeneous width but different inhomogeneous widths. It was found that the echo signals in both systems seemingly exhibit a similar variation following the increases of the optical density. However, the decay constants were different for the two systems. In the system which has a large inhomogeneous broadening, T_2 is determined uniquely from the range of the longer delay time regardless of the amount of the optical density. On the other hand, in cases with a small inhomogeneous width, the decay constant depends on the optical density. In the latter case, the correlation function defined in Eqs. (8) and (9) has a negative exponential tail provided the laser bandwidth exceeds the absorption spectral width. This reflects the Lorentzian line shape of the absorption spectrum due to the small inhomogeneous broadening. Therefore, over the wide range of the delay time, the echo temporal behavior is affected by this negative exponential tail of the correlation function. The contribution of the negative part is enhanced as the optical density of the sample is increased. However, in the opposite case with a broad inhomogeneous broadening, the echo temporal behavior is affected by this OD effect within the restricted range of the delay time corresponding to the inverse of the absorption spectral width, since the absorption spectrum has a Gaussian line profile. Therefore, in this case, T_2 is determined unambiguously at longer delay times.

The OD effect discussed above is considered to be a general phenomenon which is not restricted to the present sample of Nd:YAG. When the laser bandwidth exceeds the absorption spectral width, and especially in an optically dense sample, this effect always has to be taken into consideration. For instance, the OD effect is observable even in a sample such as Nd-doped silicate glass, which has a broad absorption band around 5850 Å whose spectral FWHM reaches about 150 cm^{-1}. In fact, the OD effect was clearly observed when the laser bandwidth was increased to 450 cm^{-1}. The details of this result will be reported elsewhere.

To summarize, the OD effect in the accumulated photon echo was investigated both experimentally and theoretically. The cause of the distortion in the temporal behavior of the echo and the appearance of the dispersion-shaped signal were ascribed to this OD effect which is caused by the change of the field correlation function as the excitation pulse propagates through the sample.

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