Accumulated photon echoes with incoherent light in Nd^{3+}-doped silicate glass

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Accumulated photon echoes were observed in Nd^{3+}-doped silicate glass by using coherent and incoherent light, and it was theoretically and experimentally verified that the time resolution is determined not by the pulse width but by the correlation time of the excitation field. Even by using a cw laser, instead of a mode-locked laser, a subpicosecond time resolution was obtained when it was operated with a broad and smooth spectrum. The broadest output spectrum, and hence the highest time resolution, was attained when the dye laser was pumped by a mode-locked Ar^+ laser, and their cavity lengths were mismatched by a few μm. In an experiment with a resolution time of 220 fsec, a new very fast decay was observed in the echo decay curve besides a slower decay already reported.

Since the first observation of accumulated photon echoes by Hesselink and Wiersma,[1] nearly transform-limited mode-locked pulse trains have been used as the excitation source, and resolution times have been longer than 1 psec, being limited by the widths of the pulses.[2-5] Although advances have been made in generating ultrashort pulses,[6] these techniques are still difficult to apply to practical uses in spectroscopy. In this paper we show theoretically and experimentally that the time resolution of the accumulated photon echoes is determined not by the pulse width but by the correlation time of the excitation field. To verify this principle we show that a subpicosecond resolution was possible even by using a cw laser (non-mode-locked) with a broad and smooth spectrum. But the highest time resolution was obtained when an incompletely mode-locked pulse train was used.

In an accumulated photon-echo experiment a sample is irradiated by two excitation beams, which are originated from a single laser. The two beams are noncollinear as in Fig. 1, and one beam is delayed with respect to the other. The excitation of the sample by the two beams results in a periodic distribution of the population in space and in frequency within the inhomogeneous width of the sample. Accumulated photon echoes can be regarded as free-induction decays induced by either of the two excitation beams from this population modulation or population grating. In Fig. 1 the directions of the accumulated photon echoes are shown when the excitation beam E_2 is delayed with respect to the other beam E_1.

Accumulated photon echoes are effective in a system where either there is a bottleneck in the optical pumping cycle, or the excited level of the resonant two levels has a long lifetime. In the former case the population grating accumulates in the ground level during the bottleneck lifetime, and reaches to a steady state. In the latter case the accumulation occurs both in the excited and the ground levels.

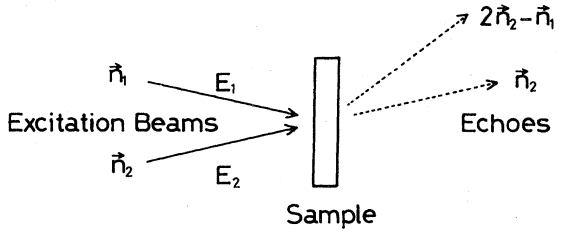

FIG. 1. Directions of the excitation beams and the accumulated photon echoes. The beam E_2 is delayed with respect to the beam E_1.

It is known that as far as the excitation intensity is within the range where the saturation broadening is negligible, the formation of the population grating can be described by a rate equation if $T_1 >> T_2$, where T_1 is the lifetime of the bottleneck or the excited level, and T_2 is the homogeneous transverse relaxation time between the two resonant levels. This condition is very well satisfied since T_1 is usually very long in the accumulated photon-echo experiment. In this situation the shape of the steady-state grating is determined simply by the power spectrum of the excitation light and the relaxation times of the sample. We express the fields of the two excitation beams as

$$E_1(t,\vec{r}) = E(t-\vec{n}_1\cdot\vec{r}/c) \ , \tag{1}$$

$$E_2(t,\vec{r}) = E(t-\tau_{12}-\vec{n}_2\cdot\vec{r}/c) \ , \tag{2}$$

where $\vec{n}_1$ and $\vec{n}_2$ are unit vectors representing the beam directions, and assumed to be nearly parallel, and τ_{12} is the delay time of the second beam E_2 with respect to the first beam E_1 at $\vec{r}=0$. The power spectrum of the excitation field is expressed as

$$S(\omega,\vec{r}) = \lim_{T\to\infty} T^{-1}\left|\int_{-T/2}^{T/2} dt[E_1(t,\vec{r})+E_2(t,\vec{r})] \times\exp(-i\omega t)\right|^2 \ . \tag{3}$$

Therefore the steady-state grating accumulated in the ground (or the excited) level is given as

$$H(\omega,\vec{r}) \propto \int_{-\infty}^{\infty} d\omega'[\gamma^2+(\omega-\omega')^2]^{-1}S(\omega',\vec{r}) \ , \tag{4}$$

where $\gamma = 1/T_2$, and the inhomogeneous width is assumed to be much larger than the spectral width of the excitation field, and the saturation of the grating is neglected.

Next, we consider a free-induction decay induced from the population grating $H(\omega,\vec{r})$ by a short pulse represented by a δ function as

$$E_3(t,\vec{r}) \propto \delta(t-\tau-\vec{n}\cdot\vec{r}/c) \ , \tag{5}$$

where $\vec{n}=\vec{n}_1$ or $\vec{n}_2$. The polarization induced by $E_3(t,\vec{r})$

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is expressed as

$$P(t,\vec{r};\tau)\propto\int_{-\infty}^{\infty}d\omega H(\omega,\vec{r})\exp[(i\omega-\gamma)(t-\tau-\vec{n}\cdot\vec{r}/c)]$$
$$=(A+B+C)\exp[-2\gamma(t-\tau-\vec{n}\cdot\vec{r}/c)]\ , \quad (6)$$

when $t\geq\tau+\vec{n}\cdot\vec{r}/c$, where

$$A=2\int_{-\infty}^{\infty}d\omega F(\omega)\exp[i\omega(t-\tau-\vec{n}\cdot\vec{r}/c)]\ , \quad (7)$$

$$B=\int_{-\infty}^{\infty}d\omega F(\omega)\exp\{i\omega[t-\tau-\tau_{12}-(\vec{n}+\vec{n}_{12})\cdot\vec{r}/c]\}\ , \quad (8)$$

$$C=\int_{-\infty}^{\infty}d\omega F(\omega)\exp\{i\omega[t-\tau+\tau_{12}-(\vec{n}-\vec{n}_{12})\cdot\vec{r}/c]\}\ , \quad (9)$$

where

$$F(\omega)=\lim_{T\to\infty}T^{-1}\left|\int_{-T/2}^{T/2}dtE(t)\exp(-i\omega t)\right|^2\ , \quad (10)$$

and $\vec{n}_{12}=\vec{n}_2-\vec{n}_1$. When $\tau_{12}>0$, the B term gives the echo polarization since the argument of the exponential function in Eq. (8) vanishes for all the frequency components at time

$$t=\tau+\tau_{12}+(\vec{n}+\vec{n}_{12})\cdot\vec{r}/c>\tau+\vec{n}\cdot\vec{r}/c\ .$$

The directions of the echo emission are, therefore, given as $\vec{n}_e'=\vec{n}_2$ when $\vec{n}=\vec{n}_1$, and $\vec{n}_e=2\vec{n}_2-\vec{n}_1$ when $\vec{n}=\vec{n}_2$. The echo along $\vec{n}_2$ interferes with the excitation beam E_2, and is observed as a heterodyne beat.[1] On the other hand, the echo along $2\vec{n}_2-\vec{n}_1$ is spatially separated from the excitation beams (see Fig. 1). We consider the latter case in the following. In the present experiment the echo is induced by $E_2(t,\vec{r})$, and thus the echo polarization is given as

$$P(t,\vec{r})=\int_{-\infty}^{t-\vec{n}_2\cdot\vec{r}/c}d\tau P(t,\vec{r};\tau)E(\tau-\tau_{12})\ , \quad (11)$$

where $P(t,\vec{r};\tau)$ is given by Eq. (6), and the relation

$$E_2(t,\vec{r})=\int_{-\infty}^{\infty}d\tau\delta(t-\tau-\vec{n}_2\cdot\vec{r}/c)E(\tau-\tau_{12})$$

is used. From Eqs. (6), (8), and (11) the echo field at an observation point $\vec{R}$ is written as

$$E_e(t,\vec{R})\propto\int_{-\infty}^{t'}d\tau G(t'-\tau-\tau_{12})\exp[-2\gamma(t'-\tau)]$$
$$\times E(\tau-\tau_{12})\ , \quad (12)$$

where $t'=t-\vec{n}_e\cdot\vec{R}/c$,

$$G(\tau)=\int_{-\infty}^{\infty}d\omega F(\omega)\exp(i\omega\tau)$$
$$=2\pi\lim_{T\to\infty}T^{-1}\int_{-T/2}^{T/2}dtE(t)E^*(t-\tau)\ , \quad (13)$$

and $G(\tau)$ is the autocorrelation function of the excitation field. In the above calculation we approximated that $\vec{n}_2\cdot\vec{r}/c\approx\vec{n}_e\cdot\vec{r}/c$. If the angle between $\vec{n}_1$ and $\vec{n}_2\approx 20$ mrad, and the beam diameter $\approx 100\,\mu$m, then $(\vec{n}_2-\vec{n}_e)\cdot\vec{r}/c\approx 6$ fsec. This is the resolution limit by the noncollinear two-beam excitation. From Eq. (12) the averaged echo intensity observed with a slow detector is given as a function of τ_{12} as

$$I(\tau_{12})\propto\int_{-\tau_{12}}^{\infty}d\tau\int_{-\tau_{12}}^{\infty}d\tau' G(\tau)G^*(\tau')G(\tau'-\tau)$$
$$\times\exp[-2(2\tau_{12}+\tau+\tau')/T_2]\ . \quad (14)$$

Equation (14) leads the following conclusion: If the width of $G(\tau)$, or the correlation time τ_c of the excitation field, is much smaller than T_2, then

$$I(\tau_{12})\propto\exp(-4\tau_{12}/T_2)\ . \quad (15)$$

If τ_c is not very short, then τ_c becomes the resolution time in measuring T_2, and the rise time of the echo intensity curve [Eq. (14)] at $\tau_{12}\approx 0$ is τ_c. Since the correlation time τ_c is related to the spectral width $\Delta\nu$ of the excitation field as $\tau_c\approx 1/\Delta\nu$, one only needs to use an excitation light with a broad and smooth spectrum to obtain a high time resolution. Therefore, it is concluded that a transform-limited ultrashort pulse train is not always necessary for the accumulated photon echo, but one can use a temporally incoherent light such as a non-transform-limited pulse train or even a cw laser with a broad and smooth spectrum. (In the case of a cw laser the term "photon echo" may no longer be appropriate.) It is usually easier to obtain a temporally incoherent light than to obtain a coherent ultrashort pulse train. The idea of a coherent transient spectroscopy in the ultrashort time region by using an incoherent light was first presented by Yajima and co-workers in the analysis of a transient four-wave mixing, and their experiment was done in Na vapor.[7] In the present paper we have treated the accumulated photon echo, where the accumulation of the population grating plays an important role. The analysis of the grating can be easily made by a rate equation,[8,9] as we have shown above, and this has clarified the relation between the spectrum of the excitation light and the time resolution.

The decay of the accumulated photon echo depends not only on the transverse relaxation but also on slower relaxations on time scales comparable to the bottleneck lifetime (for example, the spectral diffusion). To investigate the effect of the spectral diffusion on the shape of the population grating H_d, we assume the following equation:

$$\frac{\partial}{\partial t}H_d(\omega,\vec{r},t)=-(\Gamma_1+\Gamma_d)H_d(\omega,\vec{r},t)$$
$$+\int_{-\infty}^{\infty}d\omega' W(\omega'\to\omega)H_d(\omega',\vec{r},t)$$
$$+\alpha H(\omega,\vec{r})\ , \quad (16)$$

where $\Gamma_1=1/T_1$, α is the pumping rate, Γ_d is the rate of the spectral diffusion, and $W(\omega'\to\omega)$ is the probability that the transition frequency jumps from ω' to ω. The values Γ_d and $W(\omega'\to\omega)$ are related as

$$\Gamma_d=\int_{-\infty}^{\infty}d\omega' W(\omega'\to\omega)\ . \quad (17)$$

If $W(\omega'\to\omega)$ is a function of $\omega-\omega'=\Delta$, the steady-state solution of Eq. (16) is easily found, and the intensity of the echo induced from this grating is given as

$$I(\tau_{12})\propto\int_{-\tau_{12}}^{\infty}d\tau\int_{-\tau_{12}}^{\infty}d\tau' D(\tau_{12}+\tau)D^*(\tau_{12}+\tau')$$
$$\times G(\tau)G^*(\tau')G(\tau'-\tau)$$
$$\times\exp[-2(2\tau_{12}+\tau+\tau')/T_2]\ , \quad (18)$$

where $D(\theta)=1/[\Gamma_1+\Gamma_d-\tilde{W}(\theta)]$, where

$$\tilde{W}(\theta)=\int_{-\infty}^{\infty}d\Delta W(\Delta)\exp(i\theta\Delta)\ . \quad (19)$$

As an example, if $W(\Delta)$ is a Gaussian-shaped function ex-

pressed as

$$W(\Delta) \propto \exp(-\tau_d^2 \Delta^2/4) \ , \tag{20}$$

and $\tau_c \ll T_2$ and τ_d, then the echo intensity is written as

$$I(\tau_{12}) \propto \{\Gamma_1 + \Gamma_d[1-\exp(-\tau_{12}^2/\tau_d^2)]\}^{-2} \exp(-4\tau_{12}/T_2) \ . \tag{21}$$

In Fig. 2 a decay curve given by Eq. (21) is schematically shown.

A schematic diagram of the experiment is shown in Fig. 3. The excitation source was an Ar^+-laser-pumped dye laser which was operated in either mode-locked or cw (non-mode-locked) oscillation. The output was divided into two parts by a beam splitter. One of the beams (the second beam) was delayed with respect to the other (the first beam) through a variable delay line, and they were focused on a point in a sample. The average power of each beam was 10 mW at the sample, and the beam diameter at the focus was 100 μm. The angle between the two beams was 20 mrad. The sample was a 3-mm-thick piece of 3% Nd^{3+}-doped silicate glass, which was kept at about 20 K. The dye laser was resonant with the $^4I_{9/2} \leftrightarrow {}^2G_{7/2}$, $^4G_{5/2}$ transition of Nd^{3+}, and the center frequency was 5910 Å. In this case the $^4F_{3/2}$ level works as a bottleneck. The accumulated photon echoes were detected with a photomultiplier through an apperture set in the phase matching direction $2\vec{n}_2 - \vec{n}_1$. The excitation beams E_1 and E_2 were chopped at frequencies f_1 (210 Hz) and f_2 (360 Hz), respectively, and the sum frequency (f_1+f_2) component of the echo signals was picked up by a lock-in amplifier. The application of this cross-modulation technique in the accumulated photon-echo experiment resulted in a great improvement in the signal-to-noise ratio compared to the usual single-beam modulation. The echo signal was recorded by changing the delay time τ_{12} of the second beam.

In Fig. 4(a) the decay curve ot the accumulated photon-echo is shown where the excitation light was a nearly transform-limited mode-locked pulse train with pulse widths of about 3 psec. On the other hand, Fig. 4(b) is the echo decay curve when a broad-spectrum cw laser (non-mode-locked) was used. The spectral width of the cw dye laser was 14 Å corresponding to the correlation time τ_c of 0.7 psec. For the comparison of the echo decay times in the two cases, the curves are plotted in a semilog scale in Fig. 5. The echo decay curves give us the homogeneous transverse

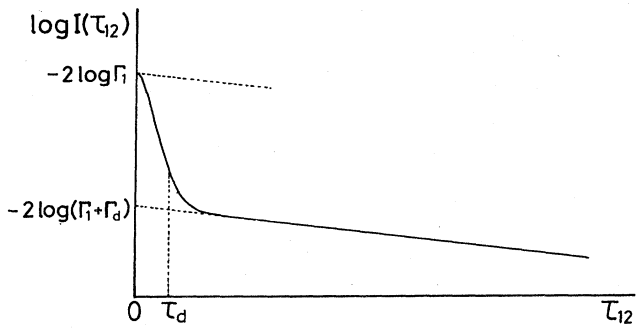

FIG. 2. The decay curve of the accumulated photon echo given by Eq. (21) where there is a spectral diffusion, and T_2 is $100\tau_d$.

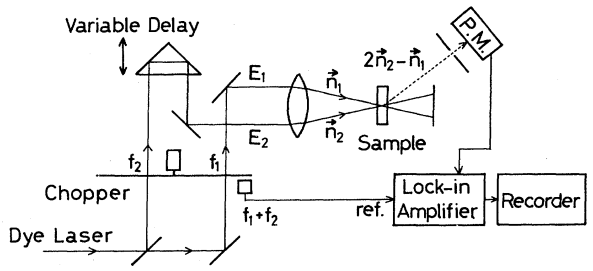

FIG. 3. Schematic diagram of the experiment.

relaxation time T_2. The two figures 5(a) and 5(b) give similar values of T_2: 60 and 57 psec, respectively, and these values are consistent with the result by Shelby.[5] The above result clearly shows that short pulses are not always necessary in the accumulated photon-echo experiment. Moreover, the fact that a higher time resolution was attained in the case of the cw laser is shown as the difference in the rise times of the curves in Figs. 4(a) and 4(b). The small humps on the decay curve in Fig. 4(b) are caused by a structure in the spectrum of the excitation light.

For a higher time resolution a laser with a broader spectrum is needed. From a practical point of view the most useful broad-spectrum laser for the accumulated photon echo experiment is an incompletely mode-locked (far from transform-limited) dye laser, which is obtained when the dye laser is pumped by a mode-locked Ar^+ laser, and their cavity lengths are mismatched by a few μm. In this way we obtained a broad-spectrum dye laser with a spectral width of 44 Å, which corresponds to the correlation time τ_c of 220 fsec, and this is the highest time resolution we attained so far. The echo decay curves obtained with this broad-spectrum laser are shown in Figs. 6 and 5(c). The peak in the curve near $\tau_{12}=0$ was observed for the first time in the present experiment. We stress here that this feature was discovered in the experiment not by using the transform-limited pulse train but by using the broad-spectrum incoherent pulse train. The width of the peak may be still

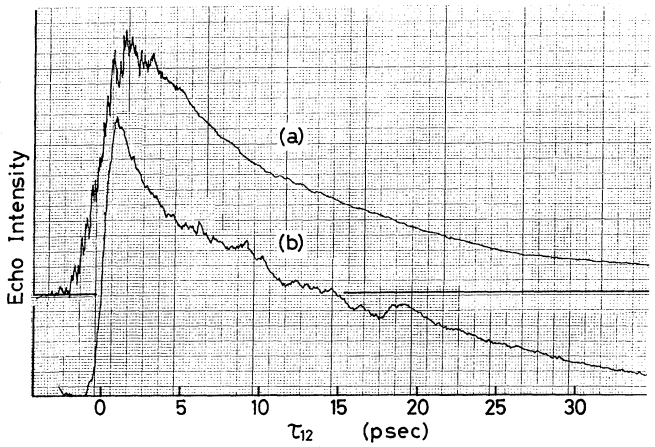

FIG. 4. The decay curves of the accumulated photon echo in Nd^{3+}-doped silicate glass where the excitation is (a) by a nearly transform-limited mode-locked pulse train, and (b) by a broad-spectrum cw laser (non-mode-locked).

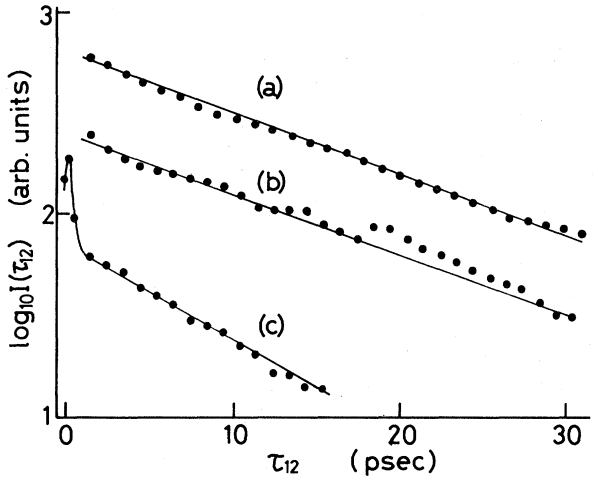

FIG. 5. Semilog plots of the echo-decay curves. Curves (a), (b), and (c) correspond to Figs. 4(a), (b), and Fig. 6, respectively.

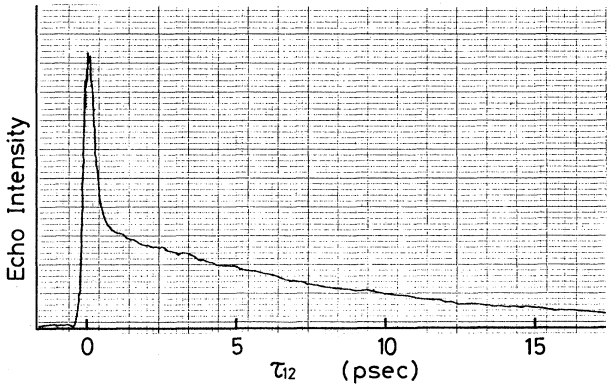

FIG. 6. The decay curve of the accumulated photon echo in Nd^{3+}-doped silicate glass by an incompletely mode-locked pulse train.

limited by the time resolution of the present experiment. A further study is needed on this point, but a possible explanation would be a spectral diffusion[10] where frequency jumps are large comparable to the inhomogeneous linewidth (see Fig. 2). The shorter T_2 obtained from the wing of the decay curve in Fig. 5(c) is due to a somewhat higher temperature of the sample than in the cases of Figs. 5(a) and 5(b).

In conclusion, we have theoretically shown that the time resolution in accumulated photon-echo experiments is determined by the correlation time of the excitation field, and verified it in the accumulated photon-echo experiment in Nd^{3+}-doped silicate glass. By using an incompletely mode-locked dye laser the highest time resolution of 220 fsec was obtained, and with this time resolution a new very fast decay in the decay curve of the accumulated photon echo was observed besides the slower decay already reported. The use of an incoherent light in accumulated photon echoes enables us to measure T_2 in femtosecond resolution without the difficulties of ultrashort pulse generation, and this method will be a useful tool in ultrafast coherent transient spectroscopy.

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