Accumulated photon echoes by using a nonlaser light source

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Accumulated photon echoes have been observed by use of a light-emitting diode. This is to our knowledge the first observation of photon echoes that are excited by a nonlaser light source. In the incoherent-light photon echo, the time resolution is equal to the inverse of the overall spectral width of the excitation light. Therefore we can easily get a high time resolution by using nonlaser light with a broad spectrum. Moreover the use of nonlaser light in the photon-echo experiment will extend the technique into wavelength regions where lasers are not currently available.

Since the first observation of photon echoes in 1964,[1] it has been believed that it is necessary to have laser pulses with pulse widths shorter than the measured dephasing time T_2. However, nearly a decade ago it was shown that by using temporally incoherent broad spectral light one can get a high time resolution equal to the inverse of the spectral width of the excitation light.[2-4]

In the incoherent-light photon echo, two beams are necessary, which are separated from a single beam by a beam splitter. The second beam is delayed by τ with respect to the first beam by an optical delay line. Therefore if we express the field of the first beam as $E_1(t) = E(t)$, that of the second beam is expressed as $E_2(t) = E(t - \tau)$. Then the field correlation function $\langle E_1(t)E_2(t)\rangle$, which is equal to $\langle E(t)E(t - \tau)\rangle$, can simply be obtained by the Fourier transform of the power spectrum of the original beam. This is why the time resolution of the incoherent-light photon echo is determined by the width of the power spectrum of the excitation light.[2]

But, in the above consideration, it is implicitly assumed that the excitation beams have a high spatial coherence and that the phase of each excitation beam is constant across the beam cross section. If the excitation beam does not have such a good spatial coherence, we need to adjust the two beams so that spatially the same coherence areas of the two beams overlap at the sample position.

In the photon-echo experiment we usually use a noncollinear configuration of the two excitation beams for spatial separation of the echo signal from the intense excitation beams. In this case the $\tau = 0$ delay line position is found by the clearest fringe pattern caused by the interference between the two beams. It is easy to find the fringe pattern if the light sources are lasers or superluminescent diodes, even when lasers are operated below threshold. But when nonlaser light sources, for example, normal light-emitting diodes (LED's), are used, it is quite difficult to find out the fringe pattern even if the beam passes through a small pinhole (e.g., 100-μm diameter). This is why, in the incoherent-light photon echo, we need temporally incoherent but spatially highly coherent light.

We have tried to find a convenient light source with a broad spectrum to use in the incoherent-light photon-echo experiment. However, light sources that have been used so far, except dye lasers, have been multimode diode lasers and superluminescent diodes.[5,6] In all these light sources, stimulated emission plays an important role, and this process from pencil-shaped gain media gives good spatial coherence to their output beams.

Recently Saikan *et al.* reported on the detection of accumulated photon echoes based on phase modulation.[7] In this method we can use collinear configuration of the two excitation beams. In this configuration, too, the $\tau = 0$ delay line position is found by the interference between the two excitation beams. However, the interference is monitored not by the spatial fringe pattern but by the brightening and darkening of the beam, which is almost uniform across the beam cross section. This way of monitoring the interference greatly simplifies the fine adjustment of the beam overlap and facilitates the use of nonlaser light in the accumulated photon echo. Moreover, in the collinear configuration of the two excitation beams we can fully utilize the high time resolution of the incoherent-light photon echo, because the ambiguity of the delay time τ, which exists in the noncollinear configuration, is absent. In this Letter we report what is to our knowledge the first observation of accumulated photon echoes excited by a nonlaser light source.

A schematic diagram of the accumulated photon-echo experiment by using an LED is shown in Fig. 1. The center wavelength, the spectral width, and the output power of the LED (Fujitsu FED073K1WA) were 740 nm, 30 nm, and 5 mW, respectively. The output beam of the LED was first focused by a lens, and in order to improve the spatial coherence of the excitation beams we used a pinhole of 100-μm diameter that transmitted ~9% of the incident power. The first beam, E_1, was phase modulated by a piezoelectric actuator at $f = 20$ kHz, and the delay time of the second beam, E_2, was changed by a stepping motor. The two beams were collinearly focused on the sample to a spot size of 0.1 mm^2 by a 5-cm focal-length lens. The power of each excitation beam at the sample was ~3.5 μW. Although the LED beam was unpolarized, the depth of the intensity modulation by the interference between the two beams was ~75% around the $\tau = 0$ delay line

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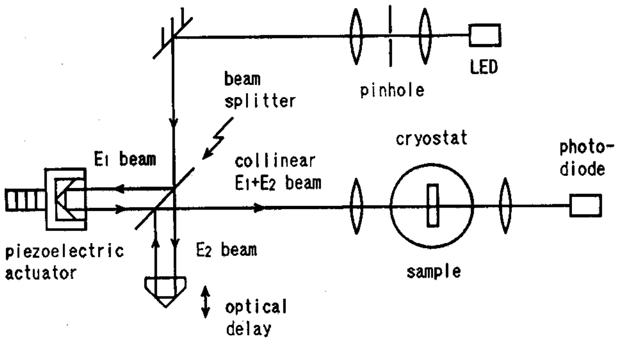
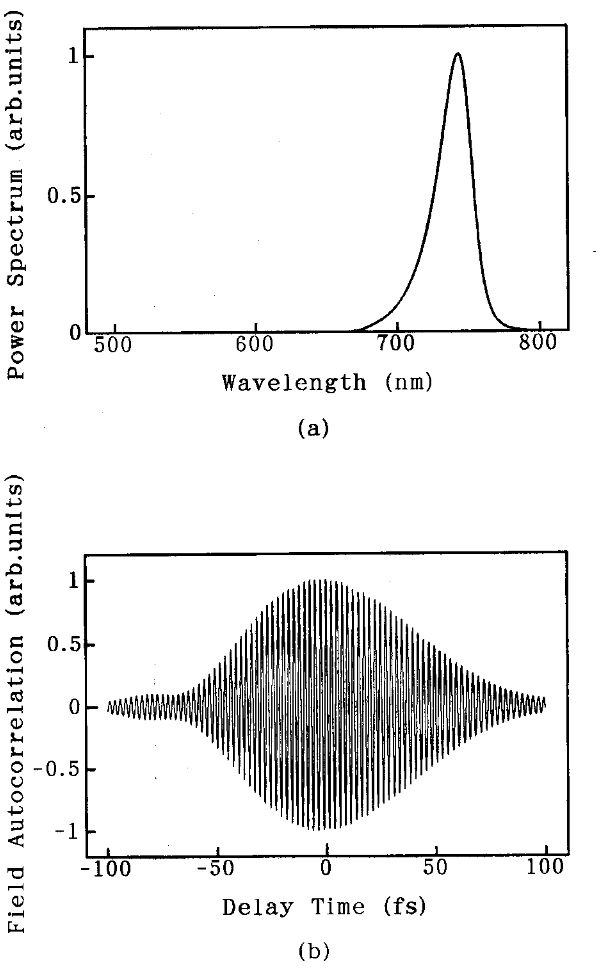


Fig. 1. Schematic diagram of the accumulated photon-echo experiment by using an LED.



(a)

(b)

Fig. 2. (a) Power spectrum and (b) field autocorrelation function of the LED.

position. The total power of the beams transmitted through the sample was detected by a photodiode and fed into a lock-in amplifier. The echo signal was obtained in the $2f$ component of the lock-in detected signal.[7]

The power spectrum of the LED is shown in Fig. 2(a). The spectrum is quite smooth, without any mode structures, unlike multimode diode lasers[5] or superluminescent diodes.[6] Fine structures in the spectrum of the excitation source result in spurious humps in the echo decay curve. It is known that the power spectrum and the field autocorrelation function have a Fourier-transformation relation with each other. We measured the field autocorrelation function $\langle E(t)E(t-\tau)\rangle$ of the LED by simply removing the sample and measuring the f component of the lock-in detected signal. The obtained curve is shown in Fig. 2(b), where the asymmetry of the curve was caused by the extra dispersion seen by the second beam, E_2, compared with that seen by the first beam, E_1, owing to the difference in the path lengths through the substrate glass of the beam splitter. The overall envelope width of the field autocorrelation function or the inverse of the spectral width is the resolution time in the incoherent-light photon-echo experiment. Therefore we can expect a resolution time of 80 fs when using the present LED.

The sample we used was 1,1′-diethyl-2,2′-quinodicarbocyanine iodide (DDI) embedded in a polyvinyl alcohol (PVA) film. It was kept at 5 K in a gas-flow cryostat (Oxford CF1204). The absorption spectrum of the sample is shown in Fig. 3, where the center wavelength of the LED is shown by an arrow. The excitation wavelength lies in the region of the inhomogeneously broadened 0–0 transition between the S_0 and S_1 levels. Like other dye-doped polymer films, the present sample (DDI in PVA) shows persistent hole burning at extremely low temperatures.[8]

The photon-echo decay curve was obtained by changing the delay time between the first beam, E_1, and the second beam, E_2. At a fixed delay time τ, the echo signal increased with time owing to the irradiation of the excitation beams, and this indicates that the signal was induced by the population grating created by persistent hole burning. We measured the echo intensity 30 s after setting each delay time. Since the spectral width of the LED was broader than the inhomogeneous width of the 0–0 transition of the sample, we did not need to erase the persistent hole that was burnt in the previous delay time.

Figure 4 shows the measured accumulated photon-echo decay curve of DDI in PVA at 5 K. In the accumulated photon-echo excited by two collinear beams, the echo decay curve is symmetric with respect to the $\tau = 0$ delay line position and decays with a time constant of $T_2/2$. The dephasing time T_2 obtained from this decay curve was 65 ps. We believe that this value of the dephasing time is reasonable when compared with those of similar samples.[9] In the present experiment we used unpolarized excitation beams; therefore the direct interference between the two beams obscured the echo signal around the $\tau = 0$

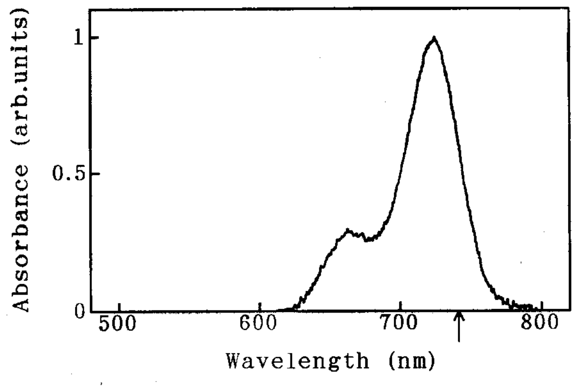


Fig. 3. Absorption spectrum of DDI in PVA.

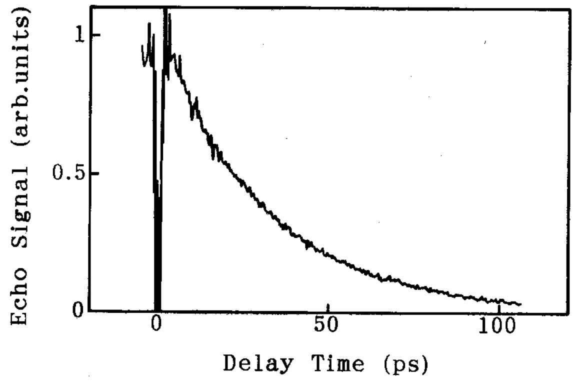


Fig. 4. Accumulated photon-echo decay curve of DDI in PVA at 5 K.

delay line position. However, one can greatly reduce this effect by using orthogonal polarizations of the two excitation beams.

In conclusion, we have succeeded in observing accumulated photon echoes by using a nonlaser light source for what is to our knowledge the first time. Since the time resolution of the incoherent-light photon echo is determined by the spectral width of the excitation source, the use of nonlaser light has great potential in the improvement of the time resolution. Moreover it will extend the photon-echo experiment into wavelength regions where lasers are not currently available.

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