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Femtosecond accumulated photon echoes excited by an incandescent lamp

Takao Fuji, Hitoshi Fukuda, Toshiaki Hattori, Hiroki Nakatsuka

Institute of Applied Physics, University of Tsukuba, Tsukuba, Ibaraki 305, Japan

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Abstract

Femtosecond accumulated photon echoes excited by an incandescent lamp have been observed. To our knowledge, this is the first observation of photon echoes excited by incoherent black-body radiation where no stimulated emission takes place. This indicates that any kind of light source can be used to excite photon echoes, a kind of nonlinear coherent transient phenomena, if the intensity is large enough or the sample has a long enough accumulation time of population grating. The black-body radiation may facilitate the extension of the possible spectral range of the photon echo experiment.

Since the first observation of photon echoes in 1964 [1], many kinds of photon echoes have been proposed and demonstrated to measure population decay time T_1, dephasing time T_2, or to monitor sublevel structures and spectral diffusion effects in the relevant two levels which are resonant with the excitation light. Because of the strong coupling between optical transition and other excitation modes such as phonons in condensed matter, the dephasing time T_2 of solids and liquids are generally very short and sometimes of the order of picoseconds or even shorter at very low temperatures. Therefore much effort has been devoted to obtain short laser pulses to excite photon echoes in condensed matter.

In 1979 Hesselink et al. observed a new type of photon-echo signal which could be excited with a low peak power laser such as a cw mode-locked dye laser. It is now called the accumulated photon echo, and the accumulation of population grating plays an important role in the echo emission [2,3]. Several years later, it was shown that by using temporally incoherent broad spectral light one can get a high time resolution which is equal to the inverse of the spectral width of the excitation light regardless of the pulse width in the measurement of T_2 in the accumulated photon echo experiment [4–6]. Since then we have tried to use various kinds of light sources to excite photon echoes such as broad spectral dye lasers [4,7], multi-mode diode lasers [8], superluminescent diodes [9], light emitting diodes [10], and synchrotron radiation [11]. However, to the best of our knowledge, there is no report on the observation of photon echoes excited by black-body radiation. The use of incoherent black-body radiation may provide a convenient extension of the wavelength range of photon echoes to the infrared and far infrared range. Moreover, it is interesting to see if photon echoes, a kind of nonlinear coherent transient phenomena, can really be excited by incoherent black-body radiation.

The dephasing process interferes with the coherent interaction between the light field and the irradiated sample, therefore by using the uncertainty relation the homogeneous width $\Delta\omega_h$ of the transition is expressed as $\Delta\omega_h = 1/(\pi T_2)$. Since, in general, the homoge-

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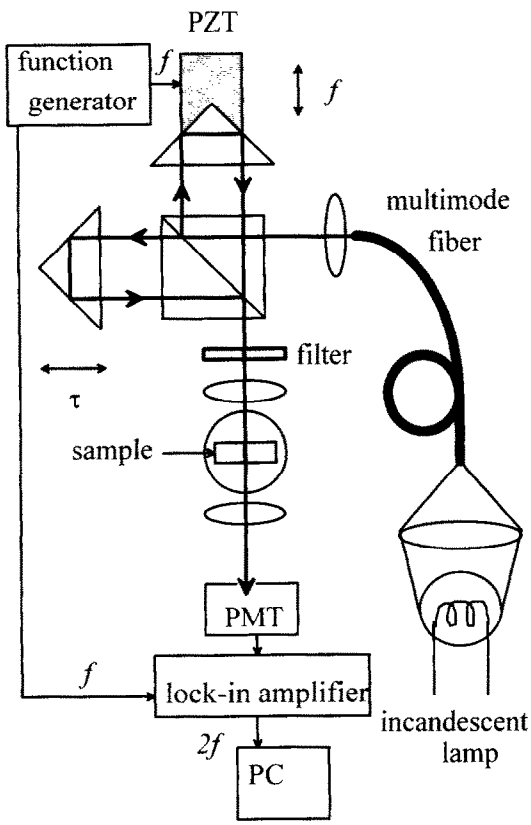

Fig. 1. Schematic diagram of the accumulated photon echo experiment using an incandescent lamp.

neous width $\Delta\omega_h$ is buried within the much broader inhomogeneous width $\Delta\omega_{ih}$, the homogeneous width $\Delta\omega_h$ or the dephasing time T_2 cannot be measured by linear-spectroscopic measurements but can only be measured by nonlinear-spectroscopic measurements such as photon echoes or hole burning.

A schematic diagram of the experimental system is shown in Fig. 1. The excitation light source was a normal type incandescent lamp whose power consumption was 650 W. The unpolarized light from the lamp was guided to a vibration isolated bench through a multi-mode optical fiber with a large core diameter of 0.9 mm. The output light from the fiber was collimated by a lens and passed through a Michelson interferometer composed of a non-polarizing beam splitter cube and two corner cube prisms. The path-length of the first excitation beam which passed through one arm of the interferometer was modulated by a piezoelectric transducer driven at f = 10 kHz, and the path-length or the delay time τ of the second excitation beam which passed through the other arm was changed by moving a corner cube prism on a translation stage. The collinear output beams or the two excitation beams from the interferometer were focused on a sample and were used to excite photon echoes. The transmitted light through the sample was detected by a photomultiplier tube, and the signal was fed into a lock-in amplifier.

The emission process of the accumulated photon echoes can be separated into two stages [2–4]. The first stage is accumulation of the population grating, or hole burning, in the inhomogeneously broadened absorption spectrum of the sample by the irradiation of the first and the second excitation beams. The second stage is scattering of the first beam as the echo by the population grating. Normally the noncollinear configuration of the two excitation beams is used in the accumulated photon echo experiment [2,3]. The first excitation beam is intensity modulated, and the echo which is emitted collinearly with the second excitation beam interferes with the second beam to produce the heterodyne signal. The heterodyne signal of the echo is detected with a lock-in amplifier.

Recently Saikan et al. developed a phase-modulation technique for accumulated photon echoes [12]. In this case one excitation beam is phase-modulated with a piezoelectric transducer as is shown in Fig. 1. In this case also it is advantageous to use noncollinear configuration of the two excitation beams. But even in the collinear configuration, if the delay time τ is well out of the region of the electric field correlation time of the excitation light, no intensity modulation occurs by the direct interference of the two excitation beams and the heterodyne signal of the echo appears in the lock-in detected signal of the $2f$ component [12]. The intensity modulation by the direct interference of the two excitation beams near τ = 0 delay line position can be greatly suppressed by using orthogonal polarization of the two excitation beams. The use of collinear configuration of the two excitation beams is inevitable when a light source with very poor spatial coherence is used to excite accumulated photon echoes. Because in this case the coherence area is much smaller than the excitation beam cross section, and we need to adjust the two excitation beams so that spatially the same coherence areas of the two beams overlap at the sample position [10].

When the inhomogeneous width of the sample is much broader than the spectral width of the excitation light, the echo intensity, $I_{\mathrm{echo}}(t,\tau)$, at time t and delay time τ is expressed as [12]

$$I_{\mathrm{echo}}(t,\tau) \propto \Re\Big[\int_0^\infty \mathrm{d}t' \{G(t'-\tau)G^*(t'-\tau) + G(t'+\tau)G^*(t'+\tau)\} \times \exp(-2t'/T_2)\, J_0(M) \exp(-iM\sin ft)\Big], \quad (1)$$

where $J_n(M)$ is the nth order Bessel function, M is the depth of the phase modulation, and G is the electric field correlation function of the excitation light which is expressed as

$$G(t'-\tau) = \left\langle \int \mathrm{d}t\, E(t)E^*(t-t'+\tau) \right\rangle_{\mathrm{av}}. \quad (2)$$

Here the period of the phase modulation, $1/f$, is assumed to be much shorter than the population grating accumulation time of the sample. In the case where the field correlation time is much shorter than the dephasing time T_2, and G can be considered to be proportional to the δ-function, Eq. (1) becomes

$$I_{\mathrm{echo}}(t,\tau) \propto \exp(-2|\tau|/T_2)\, J_0(M) \times \{J_0(M) + 2J_2(M)\cos 2ft + \ldots\}. \quad (3)$$

Therefore, the lock-in detected the $\cos 2ft$ component of the echo signal at delay time τ, $I_{\mathrm{echo}}(\tau)$, becomes

$$I_{\mathrm{echo}}(\tau) \propto \exp(-2|\tau|/T_2)\, J_0(M) J_2(M), \quad (4)$$

which is an exponentially decaying function of the delay time τ, and the decay constant is the dephasing time T_2 of the sample. In this discussion we assumed that the inhomogeneous width of the sample is much broader than the spectral width of the excitation light. But even if this condition is not satisfied, Eq. (4) is valid provided that the field correlation time of the excitation light is much shorter than T_2 [12].

On the other hand, the electric field autocorrelation of the excitation light, or the direct interference signal between the two excitation beams, can be measured by taking away the sample and detecting the lock-in signal of the $1f$ component. Fig. 2 shows the autocorrelation of the excitation light. From Fig. 2 we see

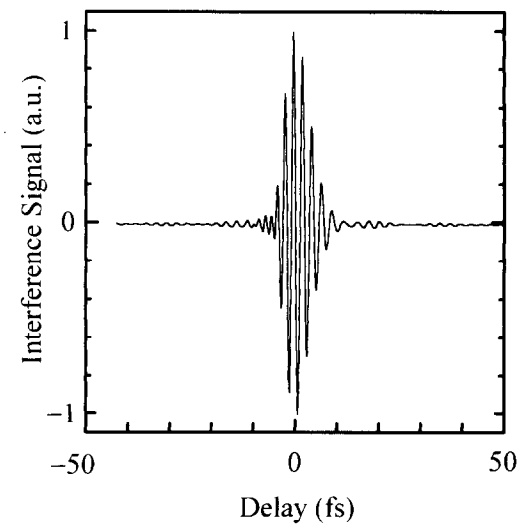

Fig. 2. Field autocorrelation of the incandescent lamp measured by the Michelson interferometer.

that the field correlation time of the excitation light is 6 fs and this is the resolution time of the photon echo experiment which is equal to the inverse of the spectral width of the excitation light [4]. The asymmetry of the autocorrelation is mainly caused by imperfect compensation of the glass dispersion between the two arms of the interferometer. In practice the effective spectral width is determined not only by the light source but also by the spectral characteristics of the optical elements in the Michelson interferometer and the photomultiplier tube (PMT).

The sample was oxazine 1 in a polyvinyl alcohol (PVA) film and was kept at 4.5 K in a gas flow cryostat (Oxford CF1204). Fig. 3 shows the absorption spectrum of oxazine 1 in PVA at 4.5 K. The 0–0 transition between the S_0 and S_1 levels is located near 675 nm at the long-wavelength side of the main peak absorption. A band-path optical filter (FWHM 25 nm, peaked at 675 nm) was inserted in front of the sample to pass only the necessary spectral component of the excitation light. The autocorrelation was measured again by inserting this filter, and it is shown in Fig. 4. The correlation time, 150 fs, is the resolution time in the present photon echo measurement. The beam intensity of the transmitted light through the filter was 0.5 μW, and the focused beam diameter at the sample was 0.4 mm.

In the present experiment we used unpolarized ex-

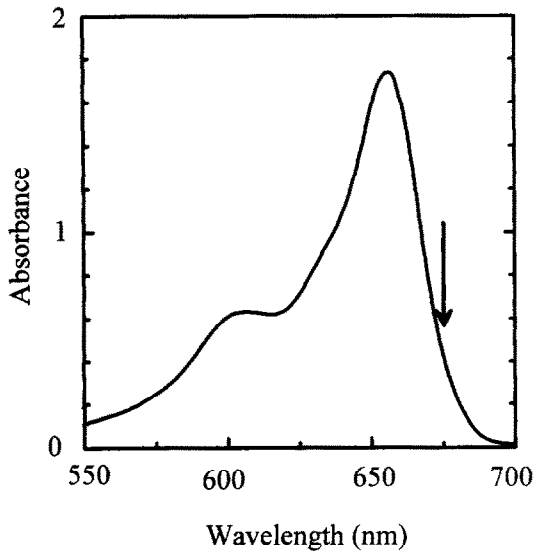

Fig. 3. Absorption spectrum of oxazine 1 in polyvinyl alcohol at 4.5 K. (The excitation wavelength of the photon echo is shown by an arrow.)

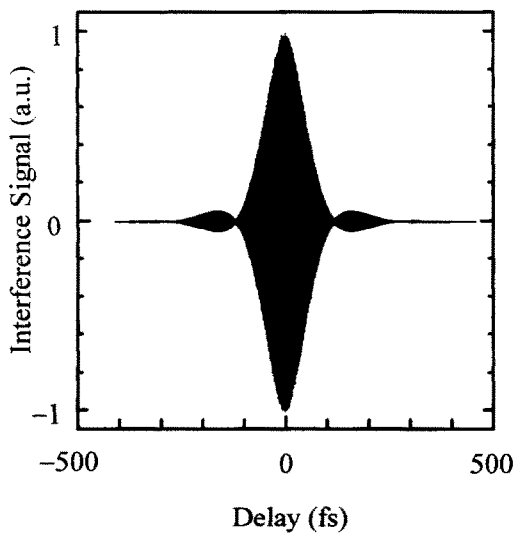

Fig. 4. Field autocorrelation of light transmitted through a filter (FWHM 25 nm, peaked at 675 nm) measured by the Michelson interferometer.

citation beams, and so the intensity modulation by the direct interference between the two excitation beams obscured the echo signal around $\tau = 0$ delay line position. Therefore we measured the echo signal in the longer delay time region where the direct interference signal of the two beams was negligible. If the delay

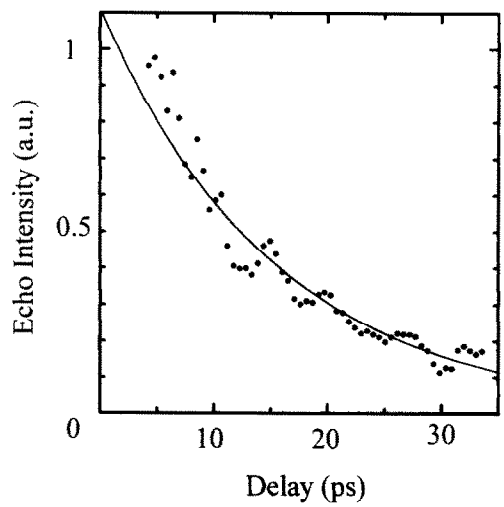

Fig. 5. Accumulated photon echo decay curve of oxazine 1 in polyvinyl alcohol at 4.5 K. The solid curve is a theoretical curve given by Eq. (4) with $T_2 = 34$ ps.

time τ was fixed, the intensity of the echo signal increased with the irradiation time. This is reasonable since the echo is considered as the scattering of the first excitation beam by the accumulated population grating created by persistent spectral hole burning [13]. The decay time of the population grating in the present sample is practically infinite at 4.5 K. At each delay time τ, the echo intensity was measured after accumulating the population grating for 10 min. A long accumulation time was necessary because the intensity of the excitation beams was quite low. In the period when the delay time was slowly changed to the next sampling point, the population grating created at the previous delay time was smeared out by the irradiation of the two excitation beams, and no echo signal was observed just after the delay time was set to the new sampling point.

The echo decay curve was obtained by changing the delay time τ by a stepping motor and is shown in Fig. 5. The solid curve is a theoretical curve given by Eq. (4) with $T_2 = 34$ ps. The obtained value of the dephasing time T_2, 34 ps, is a few times shorter than that measured by photon echoes which were excited by broad-band nanosecond laser pulses. This might be attributed to the spectral diffusion which occurred during the population grating accumulation time of 10 min. If the sample temperature is raised, $I_{\mathrm{echo}}(\tau)$ decays more rapidly with delay time τ because the

dephasing time T_2 becomes shorter for larger thermal perturbation.

In summary, we have succeeded in exciting accumulated photon echoes using an incandescent lamp or incoherent black-body radiation. If the size of the light source is reduced, we can effectively use the light intensity and reduce the time necessary for the accumulation of population grating. We are currently trying to obtain a photon echo signal using a short-arc flash lamp to minimize the necessary accumulation time of population grating.

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