cw Photon Echo

M. Mitsunaga
NTT (Nippon Telegraph and Telephone) Basic Research Laboratories, Musashino-shi, Tokyo 180, Japan

R. Kachru and E. Xu
Molecular Physics Laboratory, SRI International, Menlo Park, California 94025

M. K. Kim
Department of Physics, Wayne State University, Detroit, Michigan 48202

(Received 1 February 1989)

We apply for the first time a novel coherent transient, the cw photon echo. The propagation direction of the optical coherence prepared in a medium by a broadband cw laser beam is reversed by a pair of counterpropagating laser pulses, resulting in a continuous emission of the signal. The signal decays with a phase relaxation time T_2 and can have a quantum-beat-type modulation, giving the same wave form in one single shot as the plot of echo intensity versus pulse separation in a standard two-pulse echo experiment. Experimental demonstration is given in a Eu^{3+}:$YAlO_3$ crystal.

PACS numbers: 42.50.Md, 78.47.+p

For many years photon echoes[1] have been a powerful tool in coherent transient spectroscopy by which phase relaxation times T_2 or sublevel structures have been clarified in a number of materials.[2] In standard photon echo experiments a pair of optical pulses are applied with a time separation τ and a coherent burst of emission, the photon echo, is observed at a time τ after the second pulse. As a transient spectroscopy, most previous measurements have been made by plotting the echo intensity as a function of the delay time τ. This method, although it gives a strong signal intensity, is not performed in real time. Since a whole sequence of measurements often takes more than ten minutes, any drift of the laser intensity, wavelength, or optical alignment during the run can significantly deteriorate the data quality.

In a new approach, Mitsunaga and Brewer[3] proposed measuring T_2 in a single shot utilizing a cw laser and a pulsed laser (PL). The cw laser beam with wave vector $\mathbf{k}_1$ prepares an optical coherence in the medium and subsequently the second pulse wave vector $\mathbf{k}_2$ converts the propagation direction of the prepared coherence to $2\mathbf{k}_2-\mathbf{k}_1$. Similarly in a backward photon echo scheme[4] a pair of counterpropagating pulses ($\mathbf{k}_2$ and $-\mathbf{k}_2$) are applied, and the signal propagates along $-\mathbf{k}_1$. In both cases the signal is not a burst of emission but continuous, lasting as long as the dephasing time allows. We will call this the cw photon echo. Now, the measurement is performed *not on the echo intensity as a function of the pulse delay time, but on the echo envelope itself in real time.* This idea has already been reported and demonstrated by Schweiger *et al.*[5] in an electron-spin echo system for coherent and incoherent excitation. This Letter, we believe, is the first realization of this idea performed in the optical region. The essential difference in the optical region is that here the noncopropagating geometry plays an important role, since in this transient four-wave mixing experiment, it can be assured that the signal is generated by one photon from the cw beam and two from the PL. On the other hand, the standard copropagating geometry or electro-spin echo produces other kinds of transients,[3] yielding misleading signals.

It is worthwhile to emphasize here that the cw photon echo is distinct from the optical free induction decay[6] which also measures T_2 in real time. The latter essentially measures the hole width burned in an inhomogeneous spectrum of the medium by a narrow-band cw laser. Since the hole burning is a nonlinear process, the laser power has to be sufficiently high for a good signal-to-noise ratio. When the power is high, the hole is always power broadened, and the decay time becomes short without giving a true T_2. In the cw photon echo, however, hole burning is unnecessary at the preparation stage, and a broadband cw laser is rather favorable for exciting the whole inhomogeneous line.

The basic idea in Ref. 3 was that the photon echo obeys the superposition principle as long as the excitation intensity is low, and the theory was developed in a perturbative manner. Here we will show that by applying a broadband light source the cw echo will decay with T_2 as an exact solution even for an elevated intensity. Let us consider temporally incoherent light[7] interacting with a two-level system. The instantaneous complex Rabi frequency $\Omega(t)\equiv 2p\mathcal{E}(t)/\hbar$ for cw light is assumed to have a constant amplitude Ω_0, but to fluctuate in phase, having a stochastic nature given as

$$\langle \Omega(t)\Omega^*(t-\tau)\rangle_{\text{av}} = \Omega_0^2 e^{-\tau/\tau_c}, \tag{1}$$

where τ_c is the correlation time of the light source and gives the inverse of the laser spectral width, and the average is performed over a time much longer than τ_c. If this cw beam is applied to the two-level system, the steady-state solutions of the optical Bloch equations for the population difference w and the coherence ρ can be calculated as[8]

$$w(\Delta)=\frac{w_0(\Delta^2+1/T_{2\mathrm{eff}}^2)}{\Delta^2+1/T_{2\mathrm{eff}}^2+\Omega_0^2T_1/T_{2\mathrm{eff}}} \tag{2}$$

$$\rho(\Delta,t)=\frac{i}{2}w(\Delta)\int^t dt'\,\Omega(t')e^{-(1/T_2-i\Delta)(t-t')}, \tag{3}$$

with Δ being the detuning $\omega-\omega_0$ of the laser frequency ω from the atomic resonance frequency ω_0. The population difference in thermal equilibrium is denoted by w_0. The solution (2) is equivalent to the ordinary steady-state solution[9] except that now the ordinary dephasing time T_2 is replaced by the effective dephasing time $1/T_{2\mathrm{eff}}\equiv 1/T_2+1/\tau_c$. This leads to the quite reasonable consequence that the hole width for a weak excitation is simply the sum of the homogeneous width of the medium and the linewidth of the laser. After this preparation stage, when the counterpropagating pulses hit the sample at $t=0$, the optical coherence ρ is transferred to its complex conjugate ρ^* and starts to propagate along $-\mathbf{k}_1$.[4] By considering the contribution from the whole inhomogeneous spectrum, the signal amplitude at $t>0$ along $-\mathbf{k}_1$ is straightforwardly obtained as

$$S(t)=-\frac{i}{4}\sin\theta_f\sin\theta_b\int_{-\infty}^{\infty}d\Delta\,w(\Delta)\mathcal{G}(\Delta)\int_{-\infty}^{0}dt'\,\Omega^*(t')e^{-(t-t')/T_2+i\Delta(t+t')}, \tag{4}$$

where θ_f (θ_b) is the area of the forward (backward) pulse. The inhomogeneous distribution of the medium is represented by $\mathcal{G}(\Delta)\equiv\exp[-(\Delta/\sigma)^2]/\sqrt{\pi}\sigma$.

Two extreme cases are easily deduced from this expression. When the light source is extremely coherent ($\tau_c\rightarrow\infty$), then $\Omega(t)$ is simply a constant and $T_{2\mathrm{eff}}$ is equal to T_2. In this case the signal amplitude is

$$S(t)=-\frac{i\sqrt{\pi}w_0\Omega_0}{4\sigma}\sin\theta_f\sin\theta_b\,e^{(1+\Omega_0^2T_1T_2)/\sigma^2T_2^2}\times\frac{1+(1+\Omega_0^2T_1T_2)^{1/2}}{(1+\Omega_0^2T_1T_2)^{1/2}}e^{-t[1+(1+\Omega_0^2T_1T_2)^{1/2}]/T_2}. \tag{5}$$

As is expected, the signal decays with the inverse of the hole width including the power-broadening part. It should be pointed out here that this result presents a striking contrast to the optical free induction decay, as the signal is observed even when the saturation factor $\Omega_0^2T_1T_2$ is small compared to 1. This again shows that hole burning is unnecessary in this cw-echo scheme. On the other hand, when the laser linewidth τ_c^{-1} becomes comparable to the inhomogeneous width σ, the signal is expressed as

$$S(t)=-\frac{i\sqrt{\pi}w_0}{2\sigma}\sin\theta_f\sin\theta_b\frac{\Omega^*(-t)}{1+\Omega_0^2T_1T_{2\mathrm{eff}}}e^{-2t/T_2}. \tag{6}$$

Note that for the unsaturated regime, $\Omega_0^2T_1T_2\ll 1$, these two extreme cases coincide. In the incoherent case, however, all the higher-order terms destructively interfere and the elevated excitation intensity will not alter the decay behavior of the signal. The other important consequence is that the signal at t depends only on the cw light amplitude at $-t$. The physical interpretation of these results is as follows: The broadband cw light consists of a series of many short coherent pulses of width τ_c and completely independent of each other. Then the resulting echo signal is also a series of short independent "echolets" of width τ_c. The cw echo is like performing ordinary photon echo experiments of various delay times all at once. In other words, one is to observe *a time-reversed replica*[3,10] *of the incoherent light.* One can extend these results to a case when the system has a hyperfine structure in the ground and/or the excited state. For a weak excitation the cw photon echo is known to have an envelope modulation[11] corresponding to the hyperfine splitting frequencies. The theory mentioned here implies that the modulation should be observed with a time resolution determined by τ_c even at high intensities.

The experiment is carried out on the $^7F_0$-$^5D_0$ transition (581.6 nm) of 0.25-at.% Eu^{3+}:$\mathrm{YAlO_3}$ and is illustrated in Fig. 1. The $5\times5\times10\text{-mm}^3$ sample is cooled down to 3-30 K. A cw Ar-pumped standing-wave dye laser and a Nd-doped yttrium aluminum garnet (Nd:YAlG) pumped pulsed dye laser (PL) (5-nsec pulse width, 10-Hz repetition rate) provide cw and pulsed excitations. The correlation time of the cw laser is almost equal to the inverse of the absorption width, 6 GHz, of this crystal, which corresponds to the case of Eq. (6) in the theory. The cw beam passes an acousto-optic modulator (AOM), and provides the first pulse of any desired width. The intensities of the three pulses at the sample position are 40 mW for cw, 7.5 kW for forward PL, and

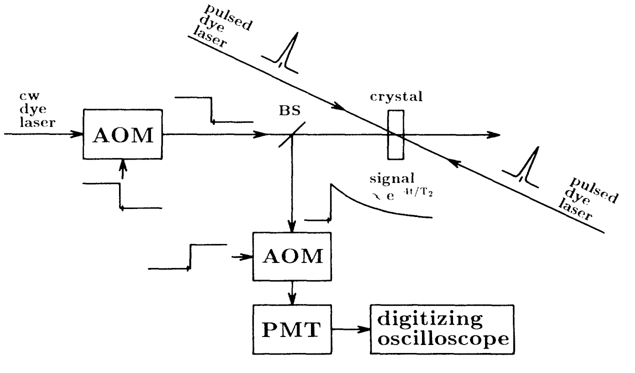

FIG. 1. Schematic of the experiment. Key: AOM, acousto-optic modulator; PMT, photomultiplier; BS, beam-splitter. Tick marks in the traces represent the time origin.

2.5 kW for backward PL. The scattered excitation light is eliminated by the AOM's in front of the photomultiplier. The signal is detected by a photomultiplier, averaged by a digitizing oscilloscope (HP 54111D), and then sent to a personal computer. Despite a small noisy signal in a single shot, it improves dramatically after averaging over about 100 times (about 1 min).

A typical cw-photon-echo signal is shown in Fig. 2(a) along with a reference signal in Fig. 2(c) where the cw beam is blocked. Once again notice that *here the echo pulse shape itself is plotted as a function of real time.* The signal envelope clearly shows a simple exponential decay with a rapid modulation on top of it. The first pulse has a width of 10 μsec, but the decay curve does not vary as a function of the pulse width. Of course, when the pulse width is reduced to less than the signal decay time, only a portion of the decay appears as the signal. In Fig. 2(b) the first pulse is externally modulated by the AOM at 10 MHz, and the echo responds with the same modulation frequency according to Eq. (6). (Multiple photon echoes of this type are reported in gas phases.[12]) The decay behavior depends strongly on the temperature. At low temperatures, the echo in this crystal is known to be very long lived and a stimulated echo can be observed even after several hours due to the long hyperfine relaxation time.[13] Hence the major part of the signal in Fig. 2(a) is considered to be the long-lived (accumulated) echo. This long-lived echo component is pronounced up to 15 K and then very quickly disappears because of the increase in the hyperfine relaxation rate, probably due to the Orbach process through the $^7F_1$ levels at 378 cm^{-1} above the ground state $^7F_0$.[14] Above 15 K, however, the cw photon echo can still be observed and here it purely represents a single-shot echo. For example, at 17 K, the decay time is 720 nsec corresponding to T_2 of 2.9 μsec. The decay time decreases gradually to about 130 nsec at 23 K and the echo finally disappears.

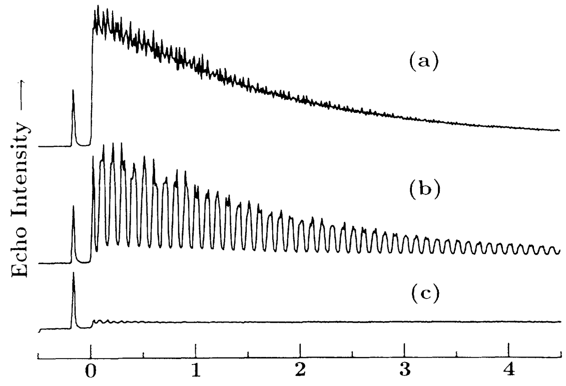

FIG. 2. cw-photon-echo wave form for the $^7F_0$-$^5D_0$ transition of Eu^{3+}:YAlO$_3$ at $T=6$ K. (a) The first pulse is a square pulse of 10-μsec width. (b) The first pulse has the same width but is amplitude modulated at 10 MHz. (c) The first pulse is blocked. Spikes at $t=-200$ nsec are the excitation pulses.

The oscillatory behavior in Fig. 2(a) is very reproducible and is pronounced at temperatures below 15 K. By Fourier transforming the signal, three frequency components of 23, 36, and 59 MHz are identified (Fig. 3). It is tempting to assign these to hyperfine splittings of this ion, which have been extensively studied by optical hole burning[15] and the rf-optical double resonance technique.[14] Of the three frequency components, 23 and 59 MHz are the two main hyperfine splittings of the ground state of the two isotopes, and 36 MHz is the difference between the two. However, the absence of other hyperfine splitting frequencies is still puzzling.

The cw photon echo has also been observed on the $^3H_4$-$^1D_2$ transition (610.5 nm) of 0.1-at.% Pr^{3+}:YAlO$_3$ at 4 K.[8] The signal exhibits a totally dissimilar wave form with an extremely deep modulation,[16] whose frequency components are identified as 7, 14, and 21 MHz of the ground-state hyperfine splittings. A separate two-pulse photon echo measurement has been performed for this crystal and an almost identical modulation pattern has been obtained. A numerical simulation based on the modulation theory also agreed with the observed decay.

Finally, an important issue has to be addressed concerning the excitation intensity dependence of T_2. As was expected from the theory described above, T_2 is recognized to be independent of the cw beam intensity. In contrast, during the course of experiments we have learned that T_2 is considerably lengthened by a reduction of the pulsed laser power. This behavior is universal for any sample we have measured, regardless of whether by the cw photon echo or the two-pulse photon echo. These observations may be related to the fact that the decay time in Fig. 2(a) shows a clear discrepancy with that obtained from photon echo experiments with a single-frequency cw laser only,[17] whose T_2 is an order of magnitude longer. This quite important fact, first pointed out in Ref. 18, will be discussed elsewhere.[19]

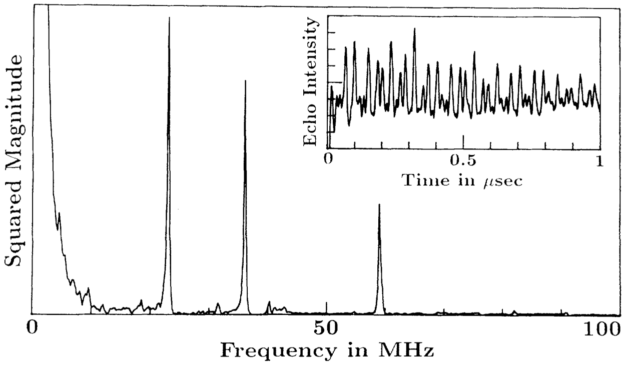

FIG. 3. Oscillatory part of the cw-photon-echo wave form (inset) and squared magnitude of its Fourier transform.

In summary, we have theoretically and experimentally demonstrated that by the combination of a cw laser and a pulsed laser one can obtain in a single shot the same echo decay curve as standard two-pulse photon echo measurements, where the plot of the echo intensity versus pulse separation is replaced by the echo pulse shape versus real time. The requirement for reproducing the decay curve is that the cw beam be weak enough to validate the superposition principle, or that the linewidth of the cw laser be broad enough to cover the inhomogeneous absorption spectrum. Specifically, in the latter case the decay rate is independent of the degree of saturation by the cw laser. In addition, this demonstration has a direct bearing on the transient optical memory.[10,20] For example, Fig. 2(b) is interpreted as the storage and retrieval of almost 50 bits of data. Finally it should be mentioned that this type of single-shot measurement can be applied to other transient four-wave mixing techniques, for example, the pump-probe method and the transient grating method, by replacing the probe pulse with a cw laser.

The comments of N. Uesugi are greatly appreciated.

---

[1] N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Phys. Rev. Lett. **13**, 567 (1964); I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. **141**, 391 (1966).

[2] See, for example, R. M. Macfarlane and R. M. Shelby, in *Spectroscopy of Solids Containing Rare Earth Ions,* edited by A. A. Kaplyanskii and R. M. Macfarlane (North-Holland, Amsterdam, 1987).

[3] M. Mitsunaga and R. G. Brewer, Phys. Rev. A **32**, 1605 (1985).

[4] M. Fujita, H. Nakatsuka, H. Nakanishi, and M. Matsuoka, Phys. Rev. Lett. **42**, 974 (1979).

[5] A. Schweiger, L. Braunschweiler, J.-M. Fauth, and R. R. Ernst, Phys. Rev. Lett. **54**, 1241 (1985); L. Braunschweiler, A. Schweiger, J.-M. Fauth, and R. R. Ernst, J. Magn. Reson. **64**, 160 (1985).

[6] R. G. Brewer and R. L. Shoemaker, Phys. Rev. A **6**, 2001 (1972); R. G. DeVoe and R. G. Brewer, Phys. Rev. Lett. **50**, 1269 (1983).

[7] S. Asaka, H. Nakatsuka, M. Fujiwara, and M. Matsuoka, Phys. Rev. A **29**, 2286 (1984); N. Morita and T. Yajima, Phys. Rev. A **30**, 2525 (1984).

[8] M. Mitsunaga (to be published).

[9] See, for example, L. Allen and J. H. Eberly, in *Optical Resonance and Two-level Atoms* (Wiley, New York, 1975).

[10] T. W. Mossberg, Opt. Lett. **7**, 77 (1982); N. W. Carlson, L. J. Rothberg, A. G. Yodh, W. R. Babbitt, and T. W. Mossberg, Opt. Lett. **8**, 483 (1983).

[11] M. Mitsunaga, K. Kubodera, and Kanbe, Opt. Lett. **11**, 339 (1986).

[12] H. Nakatsuka, S. Asaka, M. Tomita, and M. Matsuoka, Opt. Commun. **47**, 65 (1983).

[13] M. K. Kim and R. Kachru (to be published).

[14] L. E. Erickson and K. K. Sharma, Phys. Rev. B **24**, 3697 (1981).

[15] R. M. Shelby and R. M. Macfarlane, Phys. Rev. Lett. **47**, 1172 (1981).

[16] The deep modulation is characteristic of Pr^{3+} ions and has been studied in Pr^{3+}:LaF_3. Y. C. Chen, K. Chiang, and S. R. Hartmann, Opt. Commun. **29**, 181 (1979); J. B. W. Morsink and D. A. Wiersma, Chem. Phys. Lett. **65**, 105 (1979); E. A. Whittaker and S. R. Hartmann, Phys. Rev. B **26**, 3617 (1982).

[17] R. M. Shelby and R. M. Macfarlane, Phys. Rev. Lett. **45**, 1098 (1980).

[18] R. F. Liao and S. R. Hartmann, Opt. Commun. **8**, 310 (1973).

[19] R. Kachru, E. Xu, M. K. Kim, and M. Mitsunaga (to be published).

[20] M. Mitsunaga, M. K. Kim, and R. Kachru, Opt. Lett. **13**, 536 (1988).