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Incoherent Photon Echoes

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Photon echoes are generated by excitation pulses of chaotic (thermal) light. Echo signals are large and display a modulation with pulse separation due to quantum beating of hyperfine levels.

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Optical coherent transient experiments have always been performed with coherent (laser) light.[1-9] While this may be essential for experiments such as self-induced transparency and perhaps others, it is not a universal requirement. Indeed not only can optical coherent transients be generated with an incoherent lamp replacing a coherent laser, but there are decided advantages in doing so. Optical transitions can only be induced within the bandwidth of the excitation source. For a laser oscillating in at most a few well-defined modes, this means that only a small fraction of the atoms or molecules within the resonance line participate in an experiment. If in addition, successive excitations of a single resonance necessitating the use of more than one laser is required, tuning requirements are determined by the width of the lasing modes rather than the much broader width of the optical inhomogeneous or Doppler-broadened resonance line.

The key to dealing with what one might call noise-induced coherent transients is to focus attention on an isolated bandwidth $\Delta\omega = \pi/\tau$ determined by the length τ of the excitation pulse. Here $\Delta\omega$ is the full bandwidth and τ is the full length of the pulse. Within this bandwidth, the fields must be coherent no matter what their source by virtue of the uncertainty principle. Also to be noted is that in the small–pulse-area limit a coherent pulse of length τ only excites optical transitions lying within this bandwidth.[10] Successive applications of noise pulses should therefore be analyzed separately in each frequency interval $\Delta\omega = \pi/\tau$ by standard methods and the results from all distinct frequency intervals incoherently combined. We must of course respect the fact that the radiation source for each interval $\Delta\omega = \pi/\tau$ is chaotic and not coherent. The surprise is that the resulting induced radiative moment is still very large.

The practical realization of an intense tunable narrow-band thermal source is obtained by removing the output mirror from a Hänsch-type dye laser.[11] This output can be spatially filtered and further amplified to produce thermal pulses of arbitrary intensity. With use of a pair of such sources and working on the $3S$-$3P$ transition in Na vapor photon echoes were easy to generate. These echoes were generally the same size as the echoes that we observed when using laser-produced coherent excitation pulses.

In a pulse of thermal light, the relative probability of finding n photons in a single mode is given by the Bose-Einstein formula, which we use in the high-temperature limit, $\rho = \bar{n}^{-1}\exp(-n/\bar{n})$, where $\bar{n} = kT/h\nu$ is the average number of photons per mode, h is Plank's constant, $\nu = \omega/2\pi$ is the optical center frequency, k is Boltzmann's constant, and T is the temperature.[12] For a collimated beam of light of cross-sectional area A, with electric field amplitude E and duration τ, the number of photons associated with the mode whose bandwidth is $\Delta\omega = \pi/\tau$ is obtained by equating the field energy $(1/8\pi)E^2Ac\tau$ with the total photon energy $nh\nu$, where c is the velocity of light. The corresponding pulse area is θ is $\gamma E\tau = \gamma[(8\pi h\tau/A\lambda)n]^{-1/2}$ where γ is the gyroelectric ratio.

We start by restricting ourselves to a single mode. For an excitation sequence of two pulses of areas θ' and θ'' the regenerated dipole moment which gives rise to the photon echo is given by

$$p_{\text{echo}} = p_0 \sin\theta' \, (1-\cos\theta'')/2, \tag{1}$$

where p_0 is the maximum possible value of p_{echo}.[1] The radiated echo intensity is proportional to the average value of p_{echo}^2 which for incoherent excitations, incoherent also with respect to each other, we obtain by taking the product of the average value of $\sin^2\theta'$,

$$\overline{\sin^2\theta'} = \int_0^\infty \bar{n}_1^{-1} \exp(-n/\bar{n}_1)\sin^2[\theta_1(n/\bar{n}_1)^{1/2}]\,dn, \tag{2}$$

with the average value of $[\frac{1}{2}(1-\cos\theta'')]^2$,

$$\overline{[\tfrac{1}{2}(1-\cos\theta'']^2} = \int_0^\infty \bar{n}_2^{-1} \exp(-n/\bar{n}_2)\{\tfrac{1}{2}[1-\cos(\theta_2(n/\bar{n}_2)^{1/2})]\}^2\,dn. \tag{3}$$

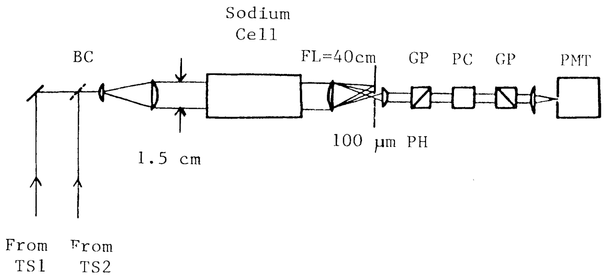

FIG. 1. Schematic diagram of our experimental setup. TS, thermal-radiation source; BC, beam combiner; PH, pinhole; GP, Glan prism; FL, focal length; and PC, Pockels-cell shutter.

We denote by θ_j the rms pulse area associated with $n=\bar{n}_j$. We find

$$\overline{\sin^2\theta'} = f(\theta_1),$$

$$\overline{[\tfrac{1}{2}(1-\cos\theta'')^2} = f(\theta_2/2) - \tfrac{1}{4}f(\theta_2),$$

where $f(\theta)=\theta\exp(-\theta^2)\int_0^\theta \exp(t^2)\,dt$. The product reaches its maximum value of $(0.64)(0.51) = 0.33 \approx \frac{1}{3}$ at $\theta_1 = 1.5 = (0.96)(\pi/2)$ and $\theta_2 = 3.1 = (0.97)(\pi)$. The average echo intensity is thus expected to be down by a factor of 3 from the intensity of a laser-induced echo. But these results only obtain outside the small-angle limit where the approximate treatment that we present does not apply. In the small-angle limit the regenerated moment Eq. (1) is linear in θ' and θ'',[2] suggesting that our results can be corrected in an approximate manner by introducing multiplicative factors η and η^2 into the terms $\sin\theta'$ and $\frac{1}{2}(1-\cos\theta'')$, respectively. The expected regenerated moment squared now becomes

$$(p_{\text{echo}}^2)_{\max} = \tfrac{1}{3}\eta^6 p_0^2, \tag{4}$$

where $\eta < 1$. As we shall see, our experimental work shows that $\eta = 0.8$.

The total radiated photon-echo signal is greater than the intensity calculated for a single mode by the ratio $\Delta\Omega/\Delta\omega$ of the resonance linewidth to the mode bandwidth. This assumes of course that the excitation pulses cover $\Delta\Omega$. The adjacent modes of the noise field by virtue of being separated by more than $\Delta\omega = \pi/\tau$ cannot interfere and must be incoherently combined. Working with a $\tau = 7$ nsec pulse and with a sample such as Na considered herein, $\Delta\Omega/2\pi = 2$ GHz whereas $\Delta\omega/2\pi = 70$ MHz, and we get an echo enhancement over that which would be attained by a single-mode laser of $(29)\frac{1}{3}\eta^6 = 10\eta^6$.

The apparatus used in the experiment is shown in Figs. 1 and 2. Except for the replacement of two dye-laser oscillators by two thermal-radiation sources (TS) (i.e., zero-point photon amplifiers) this is the apparatus that would be used in an ordinary two-pulse angled-beam echo experiment.[13] The pump beams are the frequency-doubled outputs of two Quanta-Ray DCR-1A yttrium aluminum garnet lasers. The output from the thermal-radiation sources are 7 nsec long (full width at half maximum) and have a bandwidth of approximately 10 GHz as viewed on a 14.5-GHz–free-spectral-range Fabry-Perot etalon. No stable mode structure was observed on this 10-GHz line as would be ex-

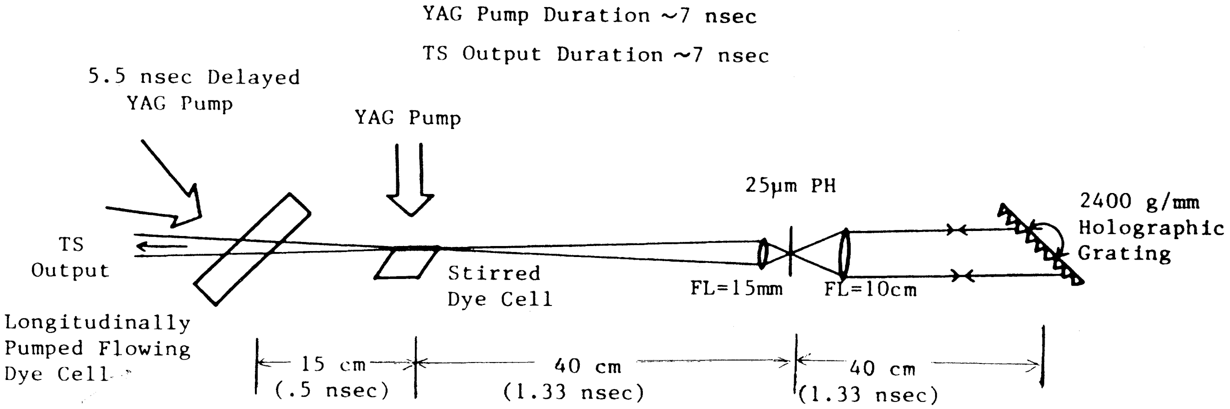

FIG. 2. Schematic diagram of the thermal-radiation sources used to generate the excitation pulses in our experiment. The dimensions were chosen so as to minimize the possibility of forming an optical cavity by spurious reflections or scatterings. The formation of such a cavity would lead to a stable mode structure in the output and destroy the chaotic (thermal) nature of the desired excitation pulses.

pected with coherent laser radiation. The central frequency of these thermal-radiation pulses can be varied within the gain curve of the dye being used (R610 in methanol) by rotation of the 2400-grooves/mm Littrow grating as indicated in Fig. 2. Tuning the two radiation sources to the resonance line is easier than if lasers had been used as now the relevant bandwidth of the radiation is 10 GHz rather than the transform-limited 70-MHz bandwidth associated with the active modes in our laser. The output pulses from the thermal-radiation sources are amplified once in a longitudinally pumped flowing-dye (R610 in methanol) cell, and finally combined at a small angle of about 0.6 mrad. The excitation pulses are then sent through a telescopic beam expander and into a sample cell approximately 10 cm long and 1.5 cm in diameter containing Na vapor. The echo, observed to emerge from the sample in the direction of $2\vec{n}_2-\vec{n}_1$, where $\vec{n}_1$ and $\vec{n}_2$ are the directions of pulses 1 and 2, respectively, is separated from the excitation pulses by use of a combination of spatial filtering and time gating of a Pockels-cell shutter. We worked on the $3^2S_{1/2}$-$3^2P_{3/2}$ transition. At a pulse separation of 60 nsec the excitation-pulse leakage signal was only one half the size of the echo signal.

Pulses 1 and 2 contain roughly 1.3×10^{12} and 8.1×10^{12} photons, respectively. These pulses are greater in intensity than the corresponding excitation pulses used with a laser source by the order of the ratio of the 10-GHz thermal bandwidth to the transform-limited 70-MHz bandwidth. At a pulse separation of 60 nsec the echoes at $(2)(60)=120$ nsec from the first excitation pulse contain roughly 3.5×10^4 photons and were obtained by optimization of the excitation pulse amplitudes and the sodium cell temperature. The latter parameter was found to be the same as that which optimized echo experiments performed with coherent laser sources replacing the thermal sources used here.

In order to make a connection with our theoretical estimates of incoherently generated echo intensity we compare the number of photons in the incoherently generated echo at an excitation-pulse separation of 60 nsec with the number of photons in a coherently (laser) generated echo under the same experimental conditions. We measured the incoherent echo intensity and determined it to contain 3.5×10^4 photons at this pulse separation. This should be compared with the number of photons in a coherently generated echo which we measured and found to be 2.5×10^4 at a pulse separation of 60 nsec. For the coherently generated echo, lasers were used that generally ran in two modes so that the number of photons per mode in this case was approximately 1.2×10^4. This gives for the echo enhancement calculated previously $10\eta^6=(3.5\times10^4)/(1.2\times10^4)\approx3$, yielding $\eta=0.8$ for our experiment. This result supports our analysis. Clearly there are more complicated processes taking place than our calculation respects and very likely an analysis that takes pulse propagation effects[14,15] and interference between neighboring modes into account is necessary. In this regard it is noteworthy that we saw no evidence of pulse breakup[2] at increased temperatures. When previously working with laser excitations we have always observed pulse breakup effects and in fact we have exploited them in the initial frequency tuning of the apparatus prior to performing an echo experiment. The absence of pulse breakup when using incoherent sources is to be expected as it is inherently due to large–pulse-area effects which we expect could not build up because of interferences from neighboring modes.

As a check on the thermal character of our excitation pulses, we generated a four-wave mixed signal from a thermal source by taking an output pulse, splitting it, optically delaying part of it, and then recombining the two parts with a small angle (~0.6 mrad) between them. The signal was then detected in the phase-matching direction $2\vec{n}_2-\vec{n}_1$, where $\vec{n}_1$ and $\vec{n}_2$ are the directions of pulses 1 and 2, respectively. This signal which was measured as a function of the delay between the pulses is displayed in Fig. 3. The corresponding four-wave mixed signal when we used an oscillator cavity in place of the thermal source always displayed a strong beating having period $2L/c$, where L was the

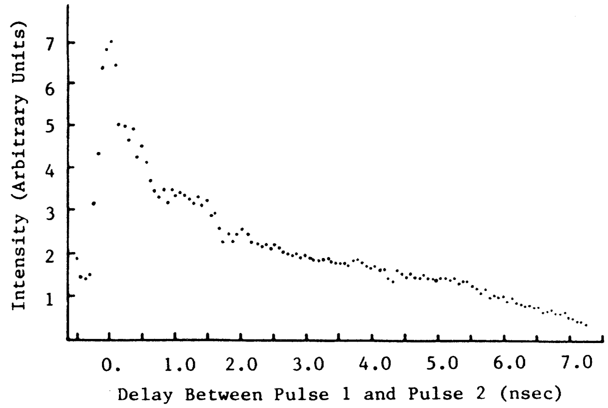

FIG. 3. Four-wave mixed signal, generated by use of one thermal source, as a function of pulse separation. The absence of any large-scale beating demonstrates that there is no stable mode structure in the thermal excitation pulses.

cavity length of the oscillator. No structure was observed with our thermal source.

The effective rise time of the four-wave mixed signal in Fig. 3 near zero delay is on the order of 100 psec. This suggests that echo experiments using incoherent excitations may be performed with a time resolution on that order.

Echo experiments in Na vapor using coherent excitations[16] have shown a weak modulated dependence of echo intensity with pulse separation and we observe the same with incoherent excitation. This is to be expected as each regenerated moment associated with each interval $\Delta\omega$ is similarly modulated.

In summary we have demonstrated that it is possible to generate photon echoes with incoherent light and achieve large signals with large single-to-noise ratios. The implication is that echo generation is possible in resonance lines of arbitrary width. Echo modulation effects due to coherent beating of the hyperfine levels are still present, so that high-resolution spectroscopy still obtains. Broadband information storage and retrieval possibilities are expanded.

Subsequent to our completing this work, related papers by Asaka *et al.*[17] and Yajima, Morita, and Ishida[18] have appeared which corroborate our findings and further demonstrate the advantages of performing coherent transient experiments with incoherent light.

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