STANDING-WAVE INDUCED BACKWARD PHOTON ECHOES IN GASES

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Recent interest in phase conjugation has made the process of degenerate-four-wave-mixing (D4WM) quite familiar.[1] In standard D4WM experiments, a standing- and travelling-wave pulse are simultaneously applied to a sample and lead to the generation of a new travelling-wave pulse. The generated pulse propagates backward with respect to, and under appropriate conditions is the phase-conjugate of the applied travelling-wave pulse. An interesting variation of this process has been suggested by Shiren.[2] He predicted that the fourth wave would be produced in inhomogeneously broadened solid-state absorbers even if the standing-wave was applied after the input travelling-wave. If the travelling- and standing-waves are displaced by the time τ, Shiren predicted that the phase-conjugate fourth wave would appear a time τ after the standing-wave. The effect predicted by Shiren displays characteristics of both D4WM and photon echoes thereby revealing the essential similarities of those effects. The echoes predicted by Shiren occur only in solids and were only recently observed.[3] A three-excitation-pulse echo process also predicted by Shiren to produce phase-conjugate output has been observed in gases.[4]

We report here a means whereby a delayed standing-wave pulse can be utilized to produce backward-propagating, potentially phase-conjugate replicas of any traditional gas-phase echo signal. Aside from its interest as a means of achieving phase conjugation, this process is also useful in relaxation studies employing echo signals because it produces echo signals unobscured by direct laser background. To produce a backward propagating replica of a gas-phase echo signal, it is only necessary to apply a standing-wave pulse, having the same frequency as the echo, at any time between the last excitation pulse and the normally produced echo signal. The stand-

ing-wave acts to "phase-conjugate" the rephasing polarization. When rephasing is complete at the normal echo time, a backward propagating echo signal is emitted.

The backward-echo-generation process reported here differs in an important way from that described by Shiren. In Shiren's case, the standing-wave acts to phase-conjugate and rephase the polarization generated in the sample material by the travelling-wave pulse. In a gas, however, the standing-wave only performs a phase-conjugation of the existing polarization. Consequently, the echo polarization must already be in the process of rephasing when the standing-wave occurs.

Working on the 2^2S-2^2P 671 nm transition of Li,[5] we have used a standing-wave to generate backward propagating replicas of well-known two-excitation-pulse photon echoes.[6] The standing-wave, applied shortly after the second travelling-wave echo excitation pulse, produced backward echoes which were easily observed when using only spatial filtering to discriminate against background signals. Importantly, standing-wave-generated backward echoes in gases can be perfectly phase-matched and perfectly rephased.

In describing the formation of echoes, it is common to assume that the excitation pulses are sufficiently brief to ensure that the entire inhomogeneous absorption profile is coherently excited. We have described standing-wave-induced-backward echoes using this approximation elsewhere.[5] In the course of our experiments, however, we have observed that the backward echo intensity depends more strongly than anticipated on the orientation of the standing-wave pulse. In an attempt to explain this effect, we have abandoned the short excitation pulse approximation, and have developed an approach to calculating the properties of echoes generated with excitation pulses of arbitrary character. As outlined below our new calculation does indeed explain our observations.

We explicitly calculate the backward echo generated in our experiments, i.e. a standing-wave is applied after two travelling-waves to produce a backward replica of the two-excitation-pulse photon echo. We assume that the spectra of each excitation pulse overlaps the inhomogeneously (i.e. Doppler) broadened absorption line of a gaseous sample of initially ground-state two-level atoms. We also assume that the sample is optically thin and neglect dispersion. The two travelling-waves (1 and 2) and the two counter-propagating components of the standing-wave (s+ and s-) all have the form

$$E_p(\vec{r},t) = 2\mathcal{E}_p(t-\eta_p)\cos[\omega_p(t-\eta_p) + \varphi_p] \quad , \tag{1}$$

where

$$\eta_p \equiv \frac{\vec{k}_p \cdot (\vec{r} - \vec{r}_0)}{\omega_p} + t_p \ . \tag{2}$$

Here $\vec{k}_p$ is the wavevector of pulse p(p = 1,2,s-,s+), t_p is the time pulse p reaches its maximum amplitude at the center of the sample $\vec{r}_0$, ω_p is the carrier frequency of pulse p, and φ_p is a constant phase. Let $\omega_\ell \equiv \omega_p$ (p = 1,2,s-,s+). We assume $\mathscr{E}_p(x)$ is maximum when x = 0, $\vec{k}_1 = \vec{k}_2 = \vec{k}$, $\vec{k}_s = \vec{k}_{s+} = -\vec{k}_{s-}$, $t_{s-} = t_{s+} = t_s$, $\mathscr{E}_{s-}(x) = \mathscr{E}_{s+}(x) \equiv \mathscr{E}_s(x)$, $\varphi_{s+} = \varphi_{s-} = \varphi_s$.

We define the Fourier transform

$$E_p^\circ(\omega) = 2\int_{-\infty}^{\infty} \mathscr{E}_p(t)\cos(\omega_\ell t + \varphi_p)e^{-i\omega t}dt \ . \tag{3}$$

The Fourier-transform seen by an atom which is moving at velocity $\vec{v}$ and is located at $\vec{r}'$ at the time t_0 is

$$E_p^a(\omega) = \left(\frac{1}{u_p}\right)e^{-i\omega s_p/u_p}E_p^\circ(\omega/u_p) \ , \tag{4}$$

where

$$u_p \equiv 1 - (\vec{k}_p \cdot \vec{v})/\omega_\ell \tag{5}$$

and

$$s_p = t_p + \vec{k}_p \cdot (\vec{r}' - \vec{v}t_0 - \vec{r}_0)/\omega_\ell \ . \tag{6}$$

Note that except for phase factors related to the time at which the atom sees the excitation pulse, $E_p^a(\omega)$ is essentially the laboratory frame Fourier transform, $E_p^\circ(\omega)$, translated in frequency by the Doppler shift.

The question now arises as to how an atom will respond to a spectrally complex laser pulse. Generally this question is difficult, but if power broadening can be neglected, it is reasonable to treat the atom as a spectral filter which only responds to frequency components within an interval of roughly $2\pi/\tau_p$ about its transition frequency. Here τ_p is the duration of the excitation pulse, and we

have assumed that the homogeneous absorption width of each atom is negligibly small. Under this assumption, the atom's response to the complex excitation pulse will be equivalent to its response to a transform-limited pulse of duration τ_p having the form[7]

$$\varepsilon_p = \left(\frac{2\pi}{\tau_p}\right) E_p^{a*}(\omega_a) e^{-i\omega_a t} + \text{c.c.} \tag{7}$$

Here ω_a is the rest frame transition frequency of the atom. The standing-wave field is simply the sum of two travelling-wave components each having the form of Eq. 7.

Using the formalism of Ref. 8, and using Eq. 7, we find that a series of three excitation pulses, the last being the standing-wave, gives rise to a polarization of the form

$$\mathcal{P}(\vec{r},t) \propto \exp\left\{-i\omega_a\left[t-(t_1-2t_2+2t_s)+\frac{\hat{k}\cdot(\vec{r}-\vec{r}_0)}{c}\right]\right\}\left\{\frac{1}{2}\left[1-J_0(\theta_0)\right]\right\}$$

$$\times\int_{-\infty}^{\infty} d\vec{v}\, g(\vec{v})\, \frac{E_1^{\circ *}(\omega_a+\vec{k}\cdot\vec{v})\, E_2^{\circ 2}(\omega_a+\vec{k}\cdot\vec{v})}{\left|E_1^{\circ}(\omega_a+\vec{k}\cdot\vec{v})\right|\left|E_2^{\circ}(\omega_a+\vec{k}\cdot\vec{v})\right|^2}\, \sin\theta_1 \sin^2(\theta_2/2)$$

$$\times \exp\left\{-i\left[\chi_s(\omega_a-\vec{k}_s\cdot\vec{v})+\chi_s(\omega_a+\vec{k}_s\cdot\vec{v})\right]\right\}$$

$$\times \exp\left\{i\omega_a\frac{\vec{v}}{c}\cdot\left[\hat{k}\left\{t-(2t_2-t_1)-\frac{\hat{k}}{c}\cdot(\vec{r}-\vec{r}_0)\right\}+\hat{k}_s\left\{\frac{2\hat{k}_s}{c}\cdot(\vec{r}-\vec{r}_0)\right\}\right]\right\}$$

$$+ \text{c.c.} \tag{8}$$

Here we have assumed for simplicity that $E_s^{\circ}(\omega) = \alpha_s e^{i\chi_s(\omega)}$ over the width of $g(\vec{v})$, where α_s is a positive constant and $g(\vec{v})$ is the velocity distribution function of the sample. Also J_0 is a Bessel's function, $\theta_0 = 8\pi d\alpha_s/\hbar$, d is the transition matrix element, $\theta_{1,2} = 4\pi d\left|E_{1,2}^{\circ}(\omega_a+\vec{k}\cdot\vec{v})\right|/\hbar$, and c is the speed of light. The polarization of Eq. 8 gives rise to the backward echo.

The integral over velocities in Eq. 8 causes the polarization to be small unless the coefficient of $\vec{v}$ in the final exponential is nearly zero. In samples sufficiently small (≈1 cm for visible ex-

citation) that we can always neglect $\omega_a v|(\vec{r}-\vec{r}_0)|/c^2$ compared to π, the coefficient is minimized at the time $t_e = 2t_2-t_1$. If we cannot neglect factors involving $\omega_a v|\vec{r}-\vec{r}_0|/c^2$, and if $\hat{k} \neq \pm \hat{k}_s$, the backward echo may not perfectly rephase. Notice that unless $t_s = (2t_2-t_1)$, the coefficient of ω_a in the first exponential factor of Eq. 8 is non-zero at the time t_e. It follows that any intrinsic rest frame frequency differences between atoms are not rephased in the backward echo. In a typical gas, however, ω_a is a constant.

In our experiment, which is described elsewhere,[5] we observed (see Fig. 1) that the echo intensity decreased sharply as the angle, β, between $\hat{k}$ and $\hat{k}_s$ was increased. The observed decrease is considerably more than can be reasonably accounted for because of changes in excitation pulse overlap geometry (solid line Fig. 1). To understand this fact in light of Eq. 8, we must first discuss the spectral character of our excitation pulses.

Our excitation pulses had a duration of $\tau_p = 6$ nsec and a spectral width of ≈ 15 Ghz. Knowing no more than this, we can only say that the Fourier components of the pulses cannot be coherent over frequency intervals wider than $\approx (2\pi/\tau_p)$. This implies that the phases of $E_1^{\circ *}(x)$, $E_2^{\circ}(x)$, and the magnitude of χ_s typically changes by a large (compared to π) random factor when x changes by $(2\pi/\tau_p)$. When $\pm\hat{k}_s = \hat{k} = \hat{z}$, only the integration over v_z is reduced by the non-constant phases of the excitation fields. If we make the approximation that the various velocity components of each atom are uncorrelated, we can estimate the additional decrease in $\mathscr{P}(\vec{r},t)$ as we increase the angle β between $\hat{k}$ and $\hat{k}_s$ from zero. Let $\hat{k}||\hat{z}$ and $\hat{k}_s$ be contained in the $\hat{z}$ - $\hat{x}$ plane. The period of phase fluctuation versus v_z will be only weakly dependent on β. However, as β is increased, phase fluctuations versus v_x will occur in intervals $\approx 2\pi/\tau_p k_s \sin\beta$. Treating the v_x integration as a random walk, we find that the v_x-dependent phase fluctuations will degrade $\mathscr{P}(r,t)$ by the factor $\sqrt{\omega_D/(2\pi/\tau_p \sin\beta)}$, where ω_D is the effective spectral width of $g(\vec{v})$, i.e. the Doppler width. Since echo intensity, I_e, is proportional to $\mathscr{P}^2(\vec{r},t)$, we find that excitation pulse incoherence

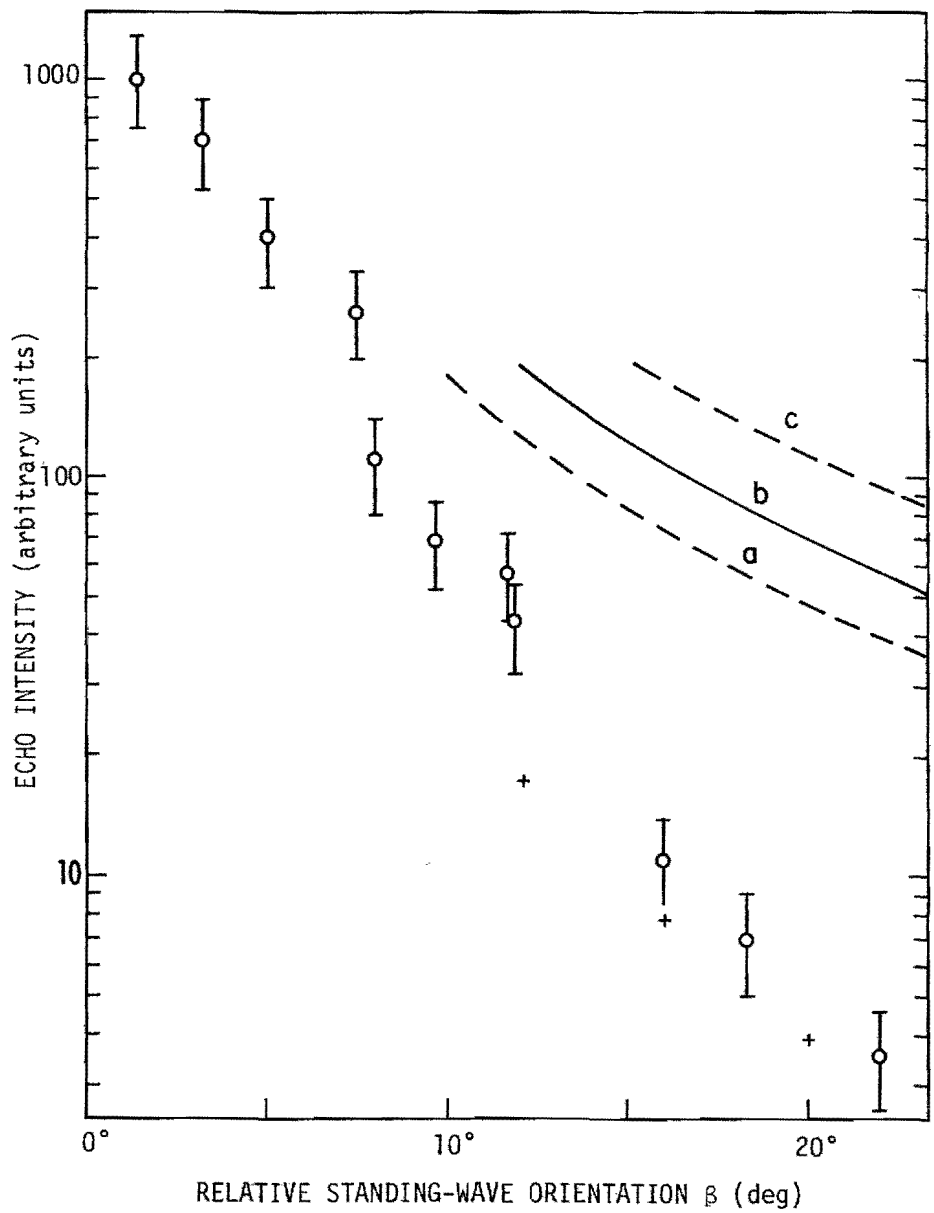

Fig. 1. Plot of Echo Intensity versus angle β. The open circles are measured data. The curves are calculated on the basis of geometric predictions. The crosses are calculated and include the effect of Eq. 9 as well as geometric effects.

leads to a decrease in echo intensity by a factor

$$\omega_D/(2\pi/\tau_p \sin\beta) \quad . \tag{9}$$

This decrease is in addition to that arising from geometric (beam overlap) effects. The three crosses shown in Fig. 1 were compiled using our experimental parameters and represent the combined effect of geometric effects and Eq. 9. Considering the simplicity of our model, the observed excellent agreement between theory and experiment must be regarded as fortuitous.

In conclusion, we have predicted and observed standing-wave-induced backward echoes. We have also described an approach to calculating echo signals which takes into account the complex spectral character of typical pulsed laser excitation.

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