

## Standing-Wave-Induced Backward Photon Echoes in Gases

N. W. Carlson and A. G. Yodh

*Division of Applied Sciences, Harvard University, Cambridge, Massachusetts 02138*

and

T. W. Mossberg

*Department of Physics, Harvard University, Cambridge, Massachusetts 02138*

(Received 7 March 1983)

It has been found that a standing-wave pulse may be employed to produce an easily detectable, backward-propagating, phase-conjugate "replica" of any traditional gas-phase photon-echo signal. Standing-wave-induced backward two-pulse photon echoes have been observed in lithium vapor while only apertures were used to block laser background. An interesting dependence of backward echo intensity on standing-wave orientation is observed and discussed.

PACS numbers: 42.65.Gv, 35.80.+s, 42.65.Bp

Photon-echo signals<sup>1-11</sup> have been shown to be very useful in the study of gas-phase relaxation processes. We report here a new class of gas-phase echo signals, which are generated by simple modification of well-known<sup>1-11</sup> echo excitation schemes so as to include a final standing-wave pulse.<sup>12</sup> The new gas-phase echo signals, *standing-wave-induced backward echoes*, are expected to be particularly valuable because they are sensitive to essentially the same relaxation processes as previously employed echo signals but are much easier to detect. Relative to "normal" echo signals generated without a standing wave, backward echoes have comparable magnitude, occur simultaneously, and propagate backward. As we will discuss below, backward echoes depend relatively weakly on standing-wave orientation; consequently, one can generally produce backward echoes unobscured by direct laser light.

A backward version of *any normal echo* signal can be produced. If the excitation laser is spectrally narrow, however, it must be tuned to a frequency at which the counterpropagating components of the standing wave interact with the same atoms. We have produced standing-wave-induced backward two-pulse photon echoes on the  $2^2S-2^2P$ , 671-nm transition of lithium (see Fig. 1). No active or passive devices other than aperture stops were used to reduce laser background.

We note that a standing-wave-induced backward echo has been predicted to occur in solids by Shiren.<sup>13</sup> In this case, an arbitrarily oriented standing-wave pulse follows a *single* traveling-wave pulse and generates an echo propagating backward with respect to the traveling wave. This process, *which does not work in Doppler-broadened media*, serves as an interesting contrast to

the process discussed here. In Shiren's case, the standing wave *induces* a macroscopic coherence to *rephase* and radiate a backward echo which can be phase conjugate to the *traveling-wave pulse*. In our case, the standing wave *modifies* the wave vector of an *already rephasing* coherence producing a backward echo which can be phase conjugate to the *original echo*. We also note that Fujita *et al.*<sup>9</sup> and Leung, Mossberg, and Hartmann<sup>14</sup> have produced gas-phase echo signals unobscured by direct laser background. However, these echoes<sup>9,14</sup> perfectly rephase only in special cases.

We show, by explicit calculation in the simple two-pulse photon-echo case, the basic mechanism

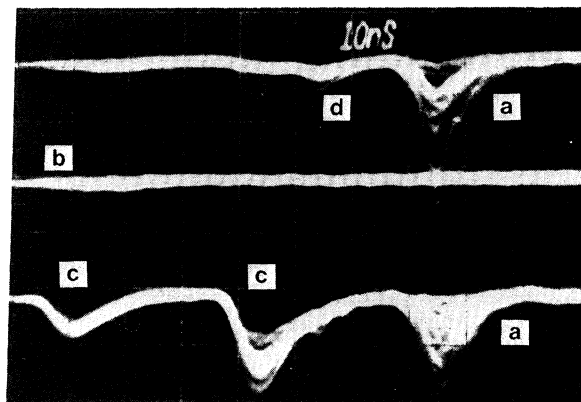


FIG. 1. Oscilloscope traces showing *a*, standing-wave-induced backward two-pulse echo; *b*, detector output with the second traveling-wave pulse blocked before the vapor region; *c*, intentionally scattered light from the two traveling-wave excitation pulses. Note that a small backward four-wave mixing signal *d* resulting from temporal overlap of the second traveling-wave and the standing-wave pulse can be seen.

responsible for backward-echo generation. A Doppler-broadened sample of ground-state atoms with central absorption frequency  $\omega_0$  is irradiated by a pulse of the form

$$\mathcal{E}_i(t)\exp[-i(\omega_0 t - \vec{k}_i \cdot \vec{r})] + \text{c.c.}$$

at the times  $t_i$  ( $i=1, 2$ ). Let  $\vec{k} \equiv \vec{k}_1 = \vec{k}_2$  and  $\tau \equiv t_2 - t_1$ . A standing-wave pulse of the form  $\mathcal{E}(t) \times \cos(\omega_0 t) \cos(\vec{k}_s \cdot \vec{r})$  is then applied at a time  $t_s$ , where  $t_2 < t_s < (t_2 + \tau)$  and  $|\vec{k}| = |\vec{k}_s|$ . Each pulse is assumed to be *sufficiently brief* that the atoms

can be regarded as stationary during the pulse. Let the sample be small compared to  $cT_2^*$ , where  $c$  is the speed of light and  $T_2^*$  is the Doppler dephasing time. Then variations of  $t_i$  and  $t_s$  throughout the sample can be neglected. For simplicity, we take pulse 1 (2) to have area  $\pi/2$  ( $\pi$ ). The first two pulses create a polarization in each atom having the form<sup>10</sup>

$$\mathcal{P}_{12} \propto d \exp(-i\omega_0 t) + \text{c.c.}, \quad (1a)$$

where

$$d \exp(-i\omega_0 t) = -\frac{1}{2} i \exp\{-i\omega_0[t - 2t_2 + t_1]\} \exp\{i\vec{k} \cdot [\vec{r} - \vec{v}(t - 2t_2 + t_1)]\}. \quad (1b)$$

Here  $\vec{r}$  and  $\vec{v}$  represent, respectively, the instantaneous position and velocity of the atom. The coefficients of  $\vec{v}$  and  $\omega_0$  vanish at  $t = 2t_2 - t_1 = t_e$  at which time  $\mathcal{P}_{12}$  is a function of  $\vec{r}$  only. One can show<sup>2,10</sup> that this  $\mathcal{P}_{12}$  gives rise to a standard two-pulse photon echo.<sup>1-7</sup> The factor  $\exp[-i(\omega_0 t - \vec{k} \cdot \vec{r})]$  indicates that the echo propagates along  $\vec{k}$ . For  $t > t_s$ , the polarization has two important terms,<sup>10,13</sup>

$$\mathcal{P}_{12s}(t > t_s) \propto d' \exp(-i\omega_0 t) + d'' \exp(-i\omega_0 t) + \text{c.c.}, \quad (2a)$$

where

$$d' = \frac{1}{2} \left[ 1 + \sum_{n=-\infty}^{\infty} (-)^n J_{2n}(\theta_s^0) \exp(2in\vec{k}_s \cdot \vec{r}_s) \right] d, \quad (2b)$$

and

$$d'' = \frac{1}{2} \left[ 1 - \sum_{n=-\infty}^{\infty} (-)^n J_{2n}(\theta_s^0) \exp(2in\vec{k}_s \cdot \vec{r}_s) \right] d^* \exp(i\omega_0 2t_s). \quad (2c)$$

Here the  $J_{2n}$  are Bessel functions,  $\theta_s^0$  is the standing-wave pulse area measured at a crest, and  $\vec{r}_s$  is the position of the atom at the time  $t_s$ .

Since the  $n \neq 0$  polarization terms of Eqs. (2b) and (2c) have wave vectors of magnitude different from  $|\vec{k}|$ , they cannot contribute to echo signals emitted at frequency  $\omega_0$ . When we neglect  $n \neq 0$  terms, Eq. (2b) becomes identical to Eq. (1b) to within an amplitude factor, and we see that echo signals still occur along  $\vec{k}$ . Again neglecting  $n \neq 0$  terms in Eq. (2c), we find that

$$d'' \exp(-i\omega_0 t) = \frac{1}{4} i \left[ 1 - J_0(\theta_s^0) \right] \exp\{-ik \cdot [\vec{r} - \vec{v}(t - 2t_2 + t_1)]\} \exp[-i\omega_0(t - 2t_s - t_1 + 2t_2)]. \quad (3)$$

We see that  $d''$  is independent of  $\vec{v}$  at  $t = t_e$ . At this time, an echo propagating as  $\exp[-i(\omega_0 t + \vec{k} \cdot \vec{r})]$ , i.e., along  $-\vec{k}$ , is emitted. If nonplane-wave excitation is considered, arguments similar to those of Ref. 13 indicate that the backward echo can be phase conjugate to the original echo. Note that the coefficient of  $\omega_0$  does not vanish at  $t = t_e$ , indicating that standing-wave-induced backward echoes as defined here will not occur in a solid (where  $\vec{v} = 0$  and  $\omega_0$  varies with position). Since the wave vector  $\vec{k}_s$  does not appear in Eq. (3), our model calculation predicts that, except for effects related to beam overlap geometry, the backward echo will be insensitive to  $\beta$ , the angle between  $\vec{k}$  and  $\vec{k}_s$ . Finally, since  $d$  in Eq. (1) may represent the polarization prior to any echo signal, an analysis similar to the one above indicates that the application of a standing-wave will "turn around" *all types of echoes*.

Despite the fact that normal [Eq. (1b)] and backward [Eq. (3)] echo polarizations are not identical, they depend on essentially the same sample parameters. Collisionally induced changes in  $\vec{v}$ ,  $\omega_0$ , or population will degrade the intensity of both types of echo. While the fine points of this degradation may in principle differ, the effect of collisions on either echo may be analyzed to yield essentially the same information.

Our experiment (see Fig. 2) utilized an excimer-laser-pumped dye laser which produced pulses of  $\approx 15$  GHz spectral and  $\tau_p = 6$  nsec temporal full width at half maximum. The pulses, which possessed a weak 10–20-nsec tail, were optically divided into the three excitation beams required, and optical delays were used to set  $t_2 - t_1 = 33$  nsec and  $t_s - t_2 = 12$  to 19 nsec. The stainless-steel lithium cell provided  $360^\circ$  optical access to

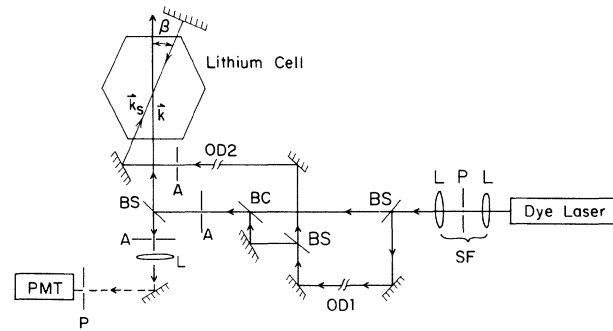


FIG. 2. Schematic of the experimental setup. BS, beam splitter; BC, beam combiner; A, aperture; L, lens; P, pinhole; OD, optical delay. OD1 determines  $t_2 - t_1$ . OD2 determines  $t_s - t_2$ . The dashed line indicates the path followed by the backward echo. The traveling-wave excitation pulses are combined at BC.

the vapor region (having a diameter  $d_v \cong 2.5$  cm). The cell was maintained at a reference temperature of  $410^\circ\text{C}$ , but we could not accurately measure the temperature of the vapor region. The excitation pulses entering the vapor cell were collimated to a diameter  $d_p = 2.7$  mm. Pulse 1 (2) had a peak power of 13 (26) W. The input component of the standing wave had a peak power of 21 W. Light emitted along  $-\hat{k}$  was detected by a photomultiplier tube. The angle  $\beta$  is easily changed without affecting detector alignment. The backward echo signal (Fig. 1) had a Li-density-optimized peak power of  $\approx 20 \mu\text{W}$  for  $\beta = 24$  mrad. When we correct for decay of the 27-nsec-life-time  $2P$  state of Li, the ratio of echo-signal peak power to first-excitation pulse (resonant fraction) peak power is  $\approx 10^{-4}$ . Normal two-pulse photon echoes generated under conditions similar to those described above, but without a standing-wave field, had a comparable magnitude.

We find that the intensity of the backward echo,  $I_e$ , decreases with increasing  $\beta$  more rapidly than can be explained on the basis of purely geometric effects (see Fig. 3). If the beam-overlap volume is contained entirely within the vapor region, i.e., if  $\beta > \beta_0 \equiv 2 \sin^{-1}(d_p/d_v)$ , we find that

$$F(\vec{v}) \equiv E_1^*(\omega_0 + \vec{k} \cdot \vec{v}) E_2^2(\omega_0 + \vec{k} \cdot \vec{v}) E_s^*(\omega_0 + \vec{k}_s \cdot \vec{v}) E_{s'}^*(\omega_0 - \vec{k}_s \cdot \vec{v}).$$

$E_x$  represents the Fourier frequency spectrum of pulse  $x$  and  $s, s'$  represent the counterpropagating components of the standing wave. If  $\vec{k} \parallel \hat{z}$  and  $\vec{k}_s$  is in the  $\hat{x}$ - $\hat{z}$  plane,  $F(\vec{v})$  is independent of  $v_y$ , but undergoes random phase variations on the scale  $\sim (k\tau_p)^{-1} [\sim (k_s\tau_p \sin\beta)^{-1}]$  as a function of  $v_z$  [ $v_x$ ]. The  $\beta$  dependence of  $f_p$  derives from the integra-

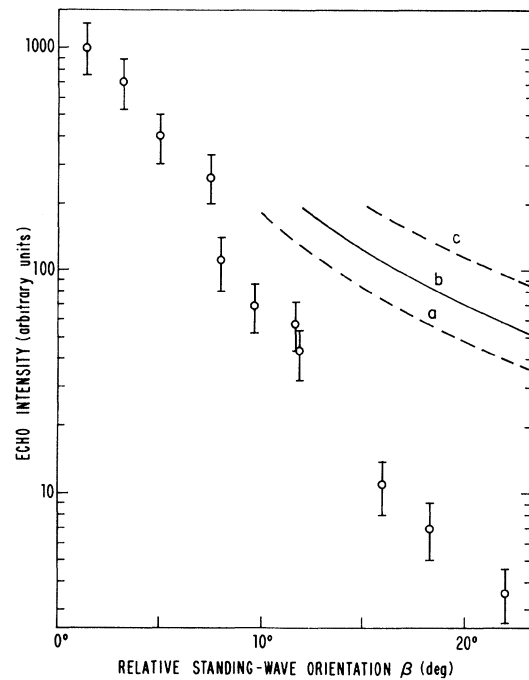


FIG. 3. Plot of echo intensity vs angle  $\beta$ . The open circles are measured data. The curves are calculated on the basis of geometric predictions (see text) with a,  $d_v = 3.1$  cm; b,  $d_v = 2.5$  cm; and c,  $d_v = 2.0$  cm.

geometric effects reduce  $I_e(\beta=0)$  by a factor  $f_g(\beta) = 3(d_p/2d_v \sin\beta)^2$ . Using our smallest  $\beta$  measurement as  $I_e(0)$ , we have plotted  $f_g(\beta > \beta_0)I_e(0)$  in Fig. 3.

In our experiment, the laser pulses are long and spectrally broad; consequently, Eq. (3), which predicts a backward echo dependent on  $\beta$  only through geometric effects, is invalid. A more general calculation, valid for arbitrarily weak excitation pulses, predicts that  $I_e(\beta) = f_p f_g I_e(0)$ , where

$$f_p(\beta) \propto \left\{ \int_0^\infty F(\vec{v}) g(\vec{v}) d^3v + \text{c.c.} \right\}^2,$$

$g(\vec{v})$  represents the velocity distribution, and, in the case of pulses such as ours whose spectral intensity is essentially constant across the Doppler profile,

tion over  $v_x$ . It can be shown that  $f_p(\beta)/f_p(0) \sim T_2^*/\tau_p \sin\beta$  for  $\beta \gg T_2^*/\tau_p$ . In our case, with  $T_2^* \sim 120$  psec,  $\tau_p \sim 6$  nsec, and  $\beta = 20^\circ$ , we have  $f_p(\beta = 20^\circ)/f_p(0) \sim 0.06$ . In fact, for  $\beta \approx 20^\circ$ , our observed echo intensity is reduced from the value expected on the basis of geometric effects by al-

most exactly this factor. However, in light of the crudeness of our calculation, this close agreement must be considered fortuitous. Note that if  $\tau_p < T_2^*$ , then  $f_p$  is independent of  $\beta$ .

We have observed standing-wave-induced backward echoes and shown the ease with which they can be detected. A strong dependence of backward echo intensity on standing-wave orientation was observed and ascribed to the complex spectral characteristics of our excitation pulses.

We thank Professor N. Bloembergen for the use of many facilities necessary to perform this work. We acknowledge the financial support of the U. S. Joint Services Electronics Program through Grant No. N00014-75-C-0648 and of the National Science Foundation through Grant No. PHY-8207080.

---

<sup>1</sup>I. D. Abella, N. A. Kurnit, and S. R. Hartmann, *Phys. Rev.* **141**, 391 (1966).

<sup>2</sup>C. K. N. Patel and R. E. Slusher, *Phys. Rev. Lett.* **20**, 1087 (1968); M. Scully, M. J. Stephen, and D. C.

Burnham, *Phys. Rev.* **171**, 213 (1968).

<sup>3</sup>P. R. Berman, J. M. Levy, and R. G. Brewer, *Phys. Rev. A* **11**, 1668 (1975); R. G. Brewer and A. Z. Genack, *Phys. Rev. Lett.* **36**, 959 (1976); R. G. Brewer and R. L. Shoemaker, *Phys. Rev. Lett.* **27**, 631 (1971).

<sup>4</sup>B. Comaskey, R. E. Scotti, and R. L. Shoemaker, *Opt. Lett.* **6**, 45 (1981).

<sup>5</sup>R. A. Forber, L. Spinelli, J. E. Thomas, and M. S. Feld, *Phys. Rev. Lett.* **50**, 331 (1983).

<sup>6</sup>R. Kachru, T. J. Chen, S. R. Hartmann, T. W. Mossberg, and P. R. Berman, *Phys. Rev. Lett.* **47**, 902 (1981).

<sup>7</sup>T. Baer, *Phys. Rev. A* **20**, 2610 (1979).

<sup>8</sup>T. Mossberg, A. Flusberg, R. Kachru, and S. R. Hartmann, *Phys. Rev. Lett.* **42**, 1665 (1979).

<sup>9</sup>M. Fujita, H. Nakatsuka, H. Nakanishi, and M. Matsuoka, *Phys. Rev. Lett.* **42**, 974 (1979).

<sup>10</sup>T. W. Mossberg and S. R. Hartmann, *Phys. Rev. A* **23**, 1271 (1981).

<sup>11</sup>R. Kachru, T. W. Mossberg, and S. R. Hartmann, *Phys. Rev. A* **21**, 1124 (1980).

<sup>12</sup>The counterpropagating traveling-wave components of the "standing wave" need not overlap in time. However, echo rephasing may degrade as the temporal separation of the traveling-wave components increases.

<sup>13</sup>N. S. Shiren, *Appl. Phys. Lett.* **33**, 299 (1978).

<sup>14</sup>K. P. Leung, T. W. Mossberg, and S. R. Hartmann, *Bull. Am. Phys. Soc.* **26**, 278 (1981).

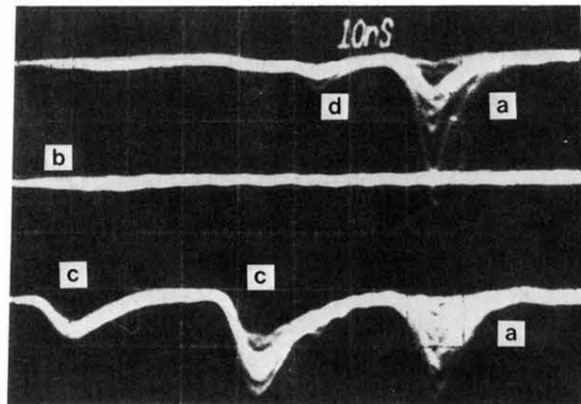


FIG. 1. Oscilloscope traces showing *a*, standing-wave-induced backward two-pulse echo; *b*, detector output with the second traveling-wave pulse blocked before the vapor region; *c*, intentionally scattered light from the two traveling-wave excitation pulses. Note that a small backward four-wave mixing signal *d* resulting from temporal overlap of the second traveling-wave and the standing-wave pulse can be seen.