Time reversal of optical pulses by four-wave mixing

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Four-wave mixing with optical pulses is considered analytically. It is shown that, whereas true time reversal of an amplitude pulse is not possible with continuous-wave pumping, this can be achieved by short-pulse pumping of a long, narrow, nonlinear medium, although time-dependent phase variations are not time reversed.

Recently there has been much interest in degenerate four-wave mixing as a mechanism for phase conjugation[1] (also known as wave-front reversal or as time reversal of monochromatic waves). Whereas monochromatic waves can be time reversed, it has been shown[2] that, when the two pump beams are continuous, this time reversal does not extend to amplitude pulses (which cannot be monochromatic by definition), although, with a thin nonlinear medium, it is possible to obtain true time reversal of time-dependent phase disturbances. In this Letter we analyze the effect of pumping a third-order nonlinear medium with pulses and show that then amplitude pulses can be time reversed.

We consider the incident pulses split up into their Fourier frequency components. Whereas in general the nonlinear interaction among any three such components from three distinct incident fields does not give a phase-matched fourth wave, we show that when the medium is pumped by counterpropagating short-pulse fields, the nonlinear interaction between these and a third, signal, field (propagating at right angles to the pulse fields) does give rise to phase-matched interaction among certain of the Fourier components and that the sum of these phase-matched components is a time-reversed counterpropagating replica of the signal-field amplitude. The principal assumptions are that the nonlinear medium is long and thin and the pump pulses are short compared with the fastest variation in signal field to be reversed accurately. The slowly varying envelope approximation is used for the pulses.

Consider three electric fields incident upon the nonlinear medium (Fig. 1): E_1 and E_2 illuminate the medium uniformly on its long side (length z_L) and E_3 the short side (width d). We expect a generated field E_4 to emerge at least approximately in the negative z direction, and we define all four fields by

$$E_1(x,t) = \tfrac{1}{2}A_1(t)e^{i(\omega t-kx)} + \text{c.c.};$$

$$A_1(t) = \int_{-\infty}^{\infty} a_1(\omega_1)e^{i\omega_1 t}\,d\omega_1, \quad \text{(1a)}$$

$$E_2(x,t) = \tfrac{1}{2}A_2(t)e^{i(\omega t+kx)} + \text{c.c.};$$

$$A_2(t) = \int_{-\infty}^{\infty} a_2(\omega_2)e^{i\omega_2 t}\,d\omega_2, \quad \text{(1b)}$$

$$E_3(z,t) = \tfrac{1}{2}A_3\left(t-\frac{z}{v}\right)e^{i(\omega t-kz)} + \text{c.c.};$$

$$A_3\left(t-\frac{z}{v}\right) = \int_{-\infty}^{\infty} a_3(\omega_3)e^{i\omega_3[(t-(z/v)]}\,d\omega_3, \quad \text{(1c)}$$

$$E_4(z,t) = \tfrac{1}{2}A_4(r,t)e^{i(\omega t+kz)} + \text{c.c.};$$

$$A_4(\mathbf{r},t) = \int_{-\infty}^{\infty} a_4(\mathbf{r},\omega_4)e^{i\omega_4 t}\,d\omega_4. \quad \text{(1d)}$$

In Eqs. (1a)–(1d) we assume the following: the medium is isotropic and nondispersive so that $k = \omega/v$, where $v = c/n$ (c is velocity of light in free space); the envelope functions A_1 and A_2 are slowly varying over the width d of the medium so that x dependence of these can be dropped inside the medium; E_3 is a pulse propagating undisturbed in the positive z direction (with no nonlinear source terms) so that its envelope function A_3 can be described by the functional form $A_3(t - z/v)$ (for simplicity the surrounding medium is assumed to have the same refractive index as the nonlinear medium, although this is not a real restriction); any diffraction or reflection of the fields is neglected.

We ignore any nonlinear refraction and assume that $|E_4| \ll |E_3| \ll |E_1|$ or $|E_2|$ (i.e., a small-signal approximation) so that the generated nonlinear polarization of interest can be expressed as

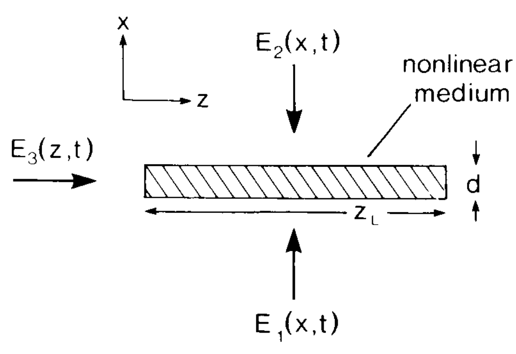

Fig. 1. Configuration of incident fields E_1, E_2, and E_3 and the nonlinear medium.

$$P^{(NL)}(\mathbf{r},t) = \tfrac{1}{2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\chi^{(3)}(\omega+\omega_1+\omega_2-\omega_3; \omega+\omega_1, \omega+\omega_2, -\omega-\omega_3)a_1(\omega_1)a_2(\omega_2) \times a_3^*(\omega_3)\exp\left\{i\left[(\omega+\omega_1+\omega_2-\omega_3)t+\left(k+\frac{\omega_3}{v}\right)z\right]\right\} \times d\omega_1 d\omega_2 d\omega_3 + \text{c.c.} \quad (2)$$

This polarization will act as the source term to generate field E_4. Substituting into the wave equation for a lossless nonmagnetic medium, assuming that all envelope functions A_1, A_2, A_3, and A_4 are slowly varying over a wavelength or period of oscillation, we obtain, on multiplying by $e^{-i(\omega+\omega_4)t}$ and integrating over time,

$$\frac{v\partial a_4}{\partial z} - i\omega_4 a_4 = i\frac{2\pi\omega}{n^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\chi^{(3)} \times (\omega+\omega_4; \omega+\omega_1, \omega+\omega_2, -\omega-\omega_1-\omega_2+\omega_4) \times a_1(\omega_1)a_2(\omega_2)a_3^*(\omega_1+\omega_2-\omega_4) \times e^{i(\omega_1+\omega_2-\omega_4)(z/v)}d\omega_1 d\omega_2. \quad (3)$$

The right-hand side of Eq. (3) is simply a function of z; x does not appear, so $a_4(\mathbf{r},\omega_4) = a_4(z,\omega_4)$, and Eq. (3) is readily solved. The origin of the coordinate system is chosen to be exactly in the middle of the nonlinear medium for convenience. For $|z| > z_L/2$, there is no nonlinear interaction and the right-hand side of Eq. (3) is zero. Therefore, for $z < z_L/2$,

$$a_4(z,\omega_4) = -i\frac{2\pi_\omega}{n^2 v}e^{i\omega_4 z/v}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\chi^{(3)}a_1 a_2 a_3^* \times \int_{(-z/2)L}^{(z/2)L} e^{i(\omega_1+\omega_2-2\omega_4)(z'/v)}dz' d\omega_1 d\omega_2. \quad (4)$$

The phase-matching integral

$$\int_{(-z/2)L}^{(z/2)L} e^{i(\omega_1+\omega_2-2\omega_4)(z'/v)}dz' = \frac{v}{(\omega_1+\omega_2-2\omega_4)}\sin\frac{(\omega_1+\omega_2-2\omega_4)z_L}{v} \quad (5)$$

can be approximated by $2\pi v\delta(\omega_1+\omega_2-2\omega_4)$ provided that either $a_1(\omega_1)$ or $a_2(\omega_2)$ is slowly varying for changes in ω_1 or $\omega_2 \sim 2\pi v/z_L$; this condition is satisfied if A_1 or A_2 (or both) is short compared with the length z_L.

Thus we have the phase-matching criterion

$$\omega_1 + \omega_2 = 2\omega_4. \quad (6)$$

From Eqs. (4) and (5) we obtain

$$a_4(z,\omega_4) = D(\omega_4)a_3^*(\omega_4)e^{i\omega_4(z/v)}, \quad (7)$$

where

$$D(\omega_4) = -i\frac{(2\pi)^2\omega}{n^2}\int_{-\infty}^{\infty}\chi^{(3)}(\omega+\omega_4; \omega+\omega_1, \omega+2\omega_4-\omega_1, -\omega-\omega_4)a_1(\omega_1)a_2(2\omega_4-\omega_1)d\omega_1. \quad (8)$$

Equation (7) shows that the frequency component $E_4(\omega + \omega_4)$ is coupled only to $E_3(\omega+\omega_4)$; this can be used to show that regardless of the form of $D(\omega_4)$, $A_4(z,t)$ is a pulse propagating in the negative z direction.

If we now assume that $\chi^{(3)}$ is a constant near ω, we may take it outside the integral in Eq. (8) and transform the convolution of a_1 and a_2 to a correlation of A_1 and A_2 using the identity

$$\int_{-\infty}^{\infty}a_1(\omega_1)a_2(2\omega_4-\omega_1)d\omega_1 = \frac{1}{2\pi}\int_{-\infty}^{\infty}A_1(t')A_2(t')e^{-2i\omega_4 t'}dt'. \quad (9)$$

We then obtain

$$A_4 = -i\frac{8\pi}{n^2 c}\omega\left[\frac{2\pi\chi^{(3)}}{n}\right]F \times \int_{-\infty}^{\infty}a_3^*(\omega_4)e^{i\omega_4[t+(z/v)-2t_p]}d\omega_4, \quad (10)$$

where we have made the additional assumptions that the product $A_1(t) \times A_2(t)$ is significant only around $t \simeq t_p$ and that its length in time is much shorter than $1/2\omega_4$ for all ω_4 of interest, so that the factor $\exp(-2i\omega_4 t')$ can be taken outside the integral in Eq. (9) as $\exp(-2i\omega_4 t_p)$; these assumptions are valid if A_1 or A_2 (or both) is a pulse that is shorter than the shortest variation in A_3 that we wish to time reverse accurately. The factor F is the correlated fluence of pulses A_1 and A_2, given by

$$F = (nc/8\pi)\int_{-\infty}^{\infty}A_1(t')A_2(t')dt'. \quad (11)$$

But, from Eq. (1c), we see that the integral in Eq. (10) is simply $A_3^*[-(t-2t_p)-(z/v)]$, so

$$A_4\left(-t-\frac{z}{v}\right) = -i\frac{8\pi\omega}{n^2 c}\left[\frac{2\pi\chi^{(3)}}{n}\right]FA_3^*\left[-(t-2t_p)-\frac{z}{v}\right]. \quad (12)$$

To interpret what this means, we can write

$$A_3(\tau) = \alpha_3(\tau)e^{i\phi_3(\tau)}, \quad (13)$$

where α_3 and ϕ_3 are real functions representing the amplitude and the phase envelopes, respectively, of the incident pulse. If $\phi_3(\tau)$ is a constant, ϕ_0, then we have

$$A_4 \propto \alpha_3\left[-(t-2t_p)-\frac{z}{v}\right]e^{-i\phi_0},$$

so that A_4 is a time-reversed replica of A_3 (subject to a constant phase shift) delayed by a time $2t_p$, which is the time taken for the relevant pulses to propagate to and from the point where the nonlinear interaction takes place. (The constant phase shift ϕ_0 is also reversed, indicating that phase conjugation may be retained under our pulsed-pump excitation.) If, however, the phase is allowed to vary in time, then

$$A_4 \propto \alpha_3\left[-(t-2t_p)-\frac{z}{v}\right]\exp\left\{-i\phi_3\left[-(t-2t_p)-\frac{z}{v}\right]\right\}.$$

Now the phase envelope has been reversed in time *and* in sense, and so the phase of the backward pulse A_4 is *not* a time-reversed replica of the input pulse A_3. These phenomena are illustrated in Fig. 2.

In terms of real fields, for an input field of the form

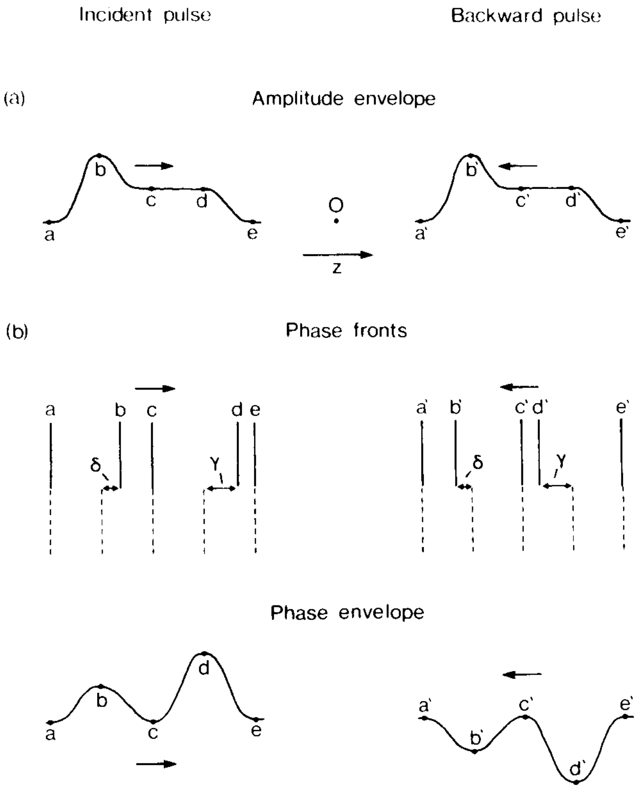

Fig. 2. Comparison of incident and backward pulses for short-pulse pumping of the nonlinear medium. (a) An observer at O sees the incident and backward amplitude envelopes as being time reversed; points a–e on the pulse pass him in the order e, d, c, b, a for the incident pulse and in the order a′, b′, c′, d′, e′ for the corresponding points on the backward pulse. (b) An observer at O sees the wavefronts a′–e′ of the backward pulse pass in the order a′, b′, c′, d′, e′, compared to the order e, d, c, b, a for the corresponding phase fronts of the incident pulse; but additionally the phase leads δ and γ have been changed to phase lags with respect to z on the backward pulse (although they remain phase leads with respect to t). The pattern of spacings between phase fronts is therefore *not* time reversed. (The phase leads and lags have been exaggerated for clarity.) The phase envelopes giving rise to these phase-front patterns show also that true time reversal does not apply because of the inversion of the backward phase envelope.

$$E_3(z,t) = \alpha_3\left(t - \frac{z}{v}\right)\cos\left[\omega t - kz + \phi_3\left(t - \frac{z}{v}\right)\right], \tag{14}$$

from Eqs. (12) and (13),

$$E_4(z,t) = \frac{8\pi\omega}{n^2 c}\left|\frac{2\pi\chi^{(3)}}{n}\right| |F| \alpha_3\left[-(t - 2t_p) - \frac{z}{v}\right] \times \cos\left\{\omega t + kz - \phi_3\left[-(t - 2t_p) - \frac{z}{v}\right] + \psi_0\right\}, \tag{15}$$

where $\psi_0 = \phi_F + \phi_x - \pi/2$ is a constant phase factor with ϕ_F and ϕ_x defined through $F = |F|\exp(i\phi_F)$ and $\chi^{(3)} = |\chi^{(3)}|\exp(i\phi_x)$ (provided that the pump pulses are not phase modulated).

Finally, in terms of intensities, we have

$$I_4\left(-t - \frac{z}{v}\right) = \left(\frac{8\pi\omega}{n^2 c}\right)^2\left[\frac{2\pi|\chi^{(3)}|}{n}\right]^2 \times |F|^2 I_3\left[-(t - 2t_p) - \frac{z}{v}\right], \tag{16}$$

which will appear time reversed even if there are time-dependent phase variations. The $\chi^{(3)}$ defined here is $6\chi_{3lmno}$ in the notation of Maker and Terhune,[3] where l, m, n, and o refer to the polarization components of E_4, E_1, E_2, and E_3, respectively. If we choose all polarizations out of the paper in Fig. 1, $\chi^{(3)} = 6\chi_{31111}$ and $2\pi\chi^{(3)}/n = 2n_2$, where n_2 is the nonlinear refractive index in electrostatic units, the additional factor of 2 arising from the nondegeneracy of all frequencies in our $\chi^{(3)}$. The factor $|F|$ is the energy per unit area of one pulse if both A_1 and A_2 are identical pulses in time with no phase modulation and they overlap optimally in the middle of the nonlinear medium.

For example, in CS_2 (where $n_2 \sim 10^{11}$ esu), ~10-psec, ~100-μJ coincident pump pulses at ~700 nm arranged to illuminate uniformly a signal beam ~500 psec (i.e., ~10 cm) long focused to ~0.03 mm (approximately the diffraction limit) in diameter should produce a backward pulse, time reversed to a resolution of $\lesssim$10 psec with an intensity, I_4, ~0.3% of the incident-signal-beam intensity, I_3. The power of the signal beam should be less than ~15 kW to prevent self-focusing and breakdown of the small-signal approximation. [In practice, the width d can be thought of as the diameter of the signal beam (here 0.3 mm) and z_L as the width of the pump beams in the z direction (here 10 cm).]

The second pump beam could be generated by reflection of the first from a mirror just underneath the signal beam in the nonlinear medium.

We have therefore demonstrated analytically that true time reversal of an amplitude pulse by four-wave mixing is possible provided that the nonlinear medium is pumped by short pulses. The fact that a backward pulse is generated even when the pump pulses are not especially short or the signal beam is continuous or $\chi^{(3)}$ is dispersive suggests possible extensions of this technique to gating, spectroscopy of $\chi^{(3)}$, or pulse correlation. A generalization of this analysis will be the subject of future work.

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