Enhanced Four-Wave Mixing by Use of Frequency-Shifted Optical Waves in Photorefractive $BaTiO_3$

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We present theory and experiments to show that the efficiency of four-wave mixing in photorefractive materials can be enhanced by use of optical waves of slightly different frequencies. The optimum frequency shift scales inversely with the response time of the material. We show that the ~1-Hz frequency shift previously observed in the output of a self-pumped phase conjugator that used photorefractive $BaTiO_3$ is the result of this inherent property of four-wave mixing.

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The time-reversed replica of an optical wave can be produced by a stimulated process in the photorefractive crystal barium titanate ($BaTiO_3$).[1] In one example the incident wave interacts by four-wave mixing with optical beams that are all self-generated and contained in the $BaTiO_3$ crystal.[2,3] The output wave is the time-reversed (or phase-conjugate) replica of the input wave. The large optical nonlinearity of photorefractive materials, which can be realized with an input-wave intensity of only a few milliwatts per centimeters,[4,5] gives this self-pumped device a high (~30%) phase-conjugate reflectivity.

A striking phenomenon was observed when the output of this self-pumped phase conjugator was allowed to feed back into a dye laser,[6] or when the self-pumped conjugator was the actual end "mirror" of a dye-laser cavity.[7] The output wavelength of the dye laser scanned slowly and monotonically over the entire gain spectrum of the dye. Wavelength scanning occurred even with a wavelength-fixing element such as an etalon, a cutoff filter, or a birefringent filter in the dye-laser cavity. Experiments revealed that the $BaTiO_3$ phase conjugator itself produced an output wave whose frequency was slightly higher or lower than the input wave by ~1 Hz.[8] This small frequency shift was compounded with each round trip inside the dye-laser cavity (~10^9 times per second) and caused the self-scanning of the dye-laser wavelength over the observed 10^{13}-Hz frequency range.[7,9] The frequency shift was caused by spontaneous translation of the photorefractive grating in the $BaTiO_3$ crystal,[7,9] although the physical origin of the grating translation was unknown.

In this Letter, we present theoretical and experimental results showing that in photorefractive materials a translating grating can have a *larger* four-wave-mixing efficiency than a stationary grating. We show that this is an inherent feature of four-wave mixing in photorefractive materials, and can occur even in the absence of a photovoltaic or intrinsic electric field.[10] We also show that the maximum efficiency occurs at *two* values of opposite sign of the grating velocity, and that the optimum velocity of the grating exhibits a power-law dependence on the total incident optical intensity. This is consistent with the observed behavior of the self-pumped phase conjugator, where the frequency shift of the output was positive or negative, depending sensitively on experimental conditions,[7] and increased with optical intensity. We propose that the self-pumped phase conjugator attains a stable steady state when its gratings translate in order to increase their four-wave-mixing efficiency.

Consider the nonlinear mixing of the four optical plane waves in a photorefractive material, as shown in Fig. 1. If the probe (4) and the reference (1) beams have different optical frequencies, they write a refractive-index grating that translates. The reading (2) beam is Doppler shifted as it scatters from the grating to produce the phase-conjugate (3) beam. The frequencies f_1 through f_4 of the four beams are related by

$$f_1 - f_4 = f_3 - f_2 \equiv \delta f \quad . \tag{1}$$

If I_1, I_2, and I_4 are the intensities of the incident waves, and I_3 is the output intensity of the phase-conjugate wave, then the diffraction efficiency of the grating $D = I_3/I_2$ is obtained by solving for the quanti-

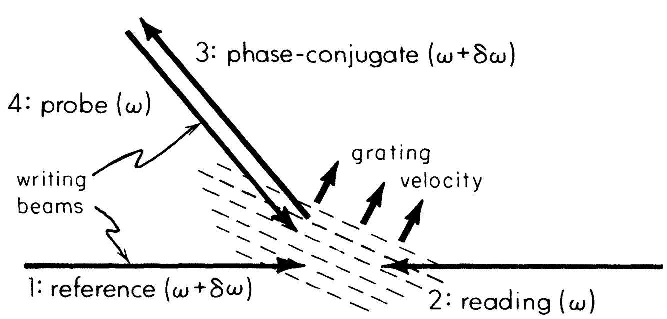

FIG. 1. Four-wave-mixing geometry. The translating grating formed by writing beams 1 and 4 diffracts reading beam 2 to produce the Doppler-shifted output phase-conjugate beam 3. The frequencies of the beams and the corresponding direction of the grating velocity are shown.

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ty X in[11]:

$$[X - I_1 I_2]\,|\Delta T + (\Delta^2 + 4X)^{1/2}|^2 + 4X|T|^2 I_2 I_4 + 4XI_4(\Delta^2 + 4X)^{1/2}\operatorname{Re}(T) = 0, \quad (2)$$

and substituting it into

$$D = \frac{4I_4 X|T|^2}{I_2|\Delta T + (\Delta^2 + 4X)^{1/2}|^2}, \quad (3)$$

where

$$T = \tanh\frac{\gamma L(\Delta^2 + 4X)^{1/2}}{2(I_1 + I_2 + I_3 + I_4)},$$

$$\Delta \equiv I_2 - I_1 - I_4,$$

and L is the length over which the waves interact. The quantity γ is the nonlinear coupling strength per unit length, and is given by[12]

$$\gamma \equiv \frac{\gamma_0}{(1 - 12\pi\tau\,\delta f)} = \frac{\gamma_0 \exp\{i\phi\}}{[1 + (2\pi\tau\,\delta f)^2]^{1/2}}. \quad (4)$$

Here $\phi \equiv \tan^{-1}(2\pi\tau\,\delta f)$ is the phase shift caused by grating motion. The response time τ depends on the grating spacing and the photoconductivity.[13] γ_0, the coupling strength per unit length for a stationary grating, is real in the absence of a photovoltaic or applied electric field.[5,12] γ_0 depends on the angles of incidence (relative to the crystal axes) and the polarizations of the waves, on material properties such as the Pockels coefficients and the photorefractive charge density, but *not* on the optical intensity (if the small dark conductivity of the crystal can be neglected).[14]

The solution of Eqs. (2)–(4) reveals that under some conditions the optical intensity of the phase-conjugate beam is largest for two nonzero values of the frequency shift δf between the two writing beams, as shown in Fig. 2. For a nonlinear coupling strength $\gamma_0 L > 2$, the curves display peaks that are symmetrically displaced on either side of $\delta f = 0$.

Physically, the enhancement of the four-wave-mixing efficiency for a nonzero frequency shift can be understood as follows: First consider the case of the writing beams having the *same* optical frequency $\delta f = 0$, so that $\phi = 0$. The phase shift between the intensity interference fringes of the writing beams and the corresponding refractive-index grating is then $\pi/2$ in photorefractive materials (if there is no applied, intrinsic, or photovoltaic electric field). The writing beams scatter with an additional $\pi/2$ phase shift from the very grating that they create, causing a net transfer of energy from one wave to the other (energy coupling).[12] If the coupling strength is large ($\gamma_0 L > 2$), energy coupling depletes one of the writing beams in a short distance. The unequal intensities of the writing beams make the modulation $m = 2(I_1 I_4)^{1/2}(I_1 + I_4)$ of the interference pattern small ($m \ll 1$) over most of the interaction region. The amplitude of the resulting refractive-index grating is proportional to the modulation, and so is correspondingly small.

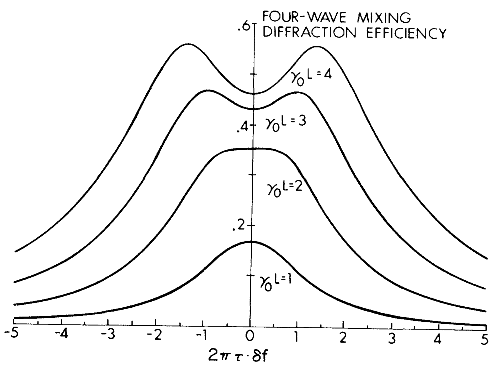

FIG. 2. Theoretical four-wave-mixing diffraction efficiency D vs the frequency shift δf between the two writing beams (in units of the response time τ of the medium), for various values of the coupling strength $\gamma_0 L$. Note the two pronounce peaks at nonzero values of δf for $\gamma_0 L > 2$.

Now consider two writing beams differing in frequency by $\delta f \neq 0$: Their interference pattern translates in the material. Because of the nonzero response time τ of the medium, the phase shift between the intensity pattern and the refractive-index grating is no longer $\pi/2$, but $\pi/2 + \phi$ [see Eq. (4)]. By decreasing the magnitude of energy coupling between the writing beams, this *increases* the average modulation of the fringes and the amplitude of the resulting photorefractive grating. Although a translating interference pattern also causes the absolute magnitude of the coupling strength to decrease, for small frequency shifts the increase in the modulation more than compensates, and the phase-conjugate signal increases. Equation (4) shows that for small frequency shifts $\delta f < 1/\tau$, the phase ϕ of γ varies nearly linearly with δf, while the magnitude $|\gamma|$ changes as $(\delta f)^2$, so that the principal effect of the grating motion is a phase shift, which increases the diffraction efficiency as shown in Fig. 2.[15] (For larger δf, the decrease in $|\gamma|$ eventually dominates, and the diffraction efficiency diminishes.)

In order to verify the enhanced four-wave-mixing efficiency of a translating photorefractive grating, experiments were performed with use of the same $BaTiO_3$ crystal as in Refs. 7 and 13. The $BaTiO_3$ crystal was in air at room temperature and had no applied electric field. An argon-ion laser was operated at 514.5 nm in a TEM_{00} mode, but in a multilongitudinal mode to reduce the coherence length of the beam to about 5 cm. The path lengths of the three incident beams were chosen so that the reading beam was not coherent with the two (mutually coherent) writing beams, thereby eliminating all but one of the possible refractive index gratings, as in Fig. 1. All of the optical beams had a diameter of ~ 0.2 mm ($1/e^2$ intensity points), and were polarized to be extraordinary rays. The probe and reference beams made angles in air of 155° and 102°, respectively, with the direction of the crystal's positive c axis. By using this beam geometry and polarization, we took advantage of the large Pockels coefficient r_{42} of $BaTiO_3$ to obtain a large coupling strength. The frequency of the writing beam 1 (see Fig. 1) was Doppler shifted a few hertz by reflection from a piezoelectrically controlled prism moving at constant velocity. In the four-wave-mixing experiments, a reading beam was carefully aligned to be counterpropagating to the reference beam. Feedback into the laser was minimized by a Faraday optical isolator between the laser and the rest of the apparatus.

The coupling strength for a stationary grating $\gamma_0 L$ was determined by blocking the reading beam and measuring the magnitude of the energy coupled between the two writing beams (1 and 4) when they had an incident power ratio of $1{:}10^6$. The power of the weak beam (1) transmitted through the crystal was observed to increase by a factor of ~ 650, giving a coupling strength of $\gamma_0 L = \frac{1}{2}\ln(650) = 3.2$.

The writing beams in the four-wave mixing experiments had comparable incident powers ($P_1 = 1.15$ mW, $P_4 = 1.25$ mW). The reading beam power ($P_2 = 0.031$ mW) was made deliberately weak to prevent it from initiating self-pumped phase conjugation.[2] Figure 3 shows the experimentally determined four-wave-mixing signal as a function of the frequency shift δf between the two writing beams. Note that the maximum four-wave-mixing efficiency occurs for nonzero frequency shifts, as predicted by the theoretical curves.

The data of Fig. 3 show that the four-wave-mixing efficiency is slightly asymmetric around $\delta f = 0$. This is due to a slight mismatch between the Gaussian wavefront curvatures of the two counterpropagating beams 1 and 2. By varying the curvatures, the asymmetry of the data can be skewed in either direction.[16] The asymmetry does not occur when the two beams have matching curvatures and so form a phase-conjugate pair.

The energy coupled between the same two writing beams was measured with the reading beam blocked, and, as expected from theory,[17] was a monotonically decreasing function of frequency shift, as shown in Fig. 4. The symmetry of these data about $\delta f = 0$ implies that no significant photovoltage or intrinsic uniform electric field exists in barium titanate.[5,12]

In successive sets of data in Figs. 3 and 4 the response time of the material was decreased by successively doubling the powers of all of the beams (while keeping the ratio of the beam powers unchanged). The four-wave-mixing results show that the optimum frequency shift increased as the response time was decreased. In Fig. 5 are log-log plots of two separate measures of the responsible time—the optimum frequency shift for four-wave mixing $(\delta f)_{\rm FWM}$, and the half-width at half maximum for energy coupling $(\delta f)_{\rm EC}$—versus total power. These plots have an average slope of 0.61 ± 0.06, in good agreement with a more direct measurement of the scaling of response time with intensity in this same $BaTiO_3$ crystal,[13] which gave $1/\tau \sim I^{0.65}$.

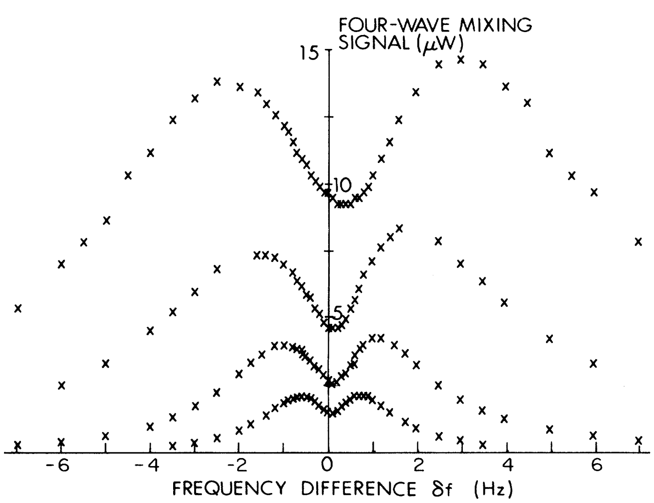

FIG. 3. Experimental four-wave-mixing signal vs the frequency shift δf between the two writing beams, for four different values of the response time of the $BaTiO_3$ crystal. The coupling strength was $\gamma_0 L = 3.2$. The total incident power for successively higher curves is in the ratio 1:2:4:8. Note the two pronounced peaks for nonzero frequency shifts.

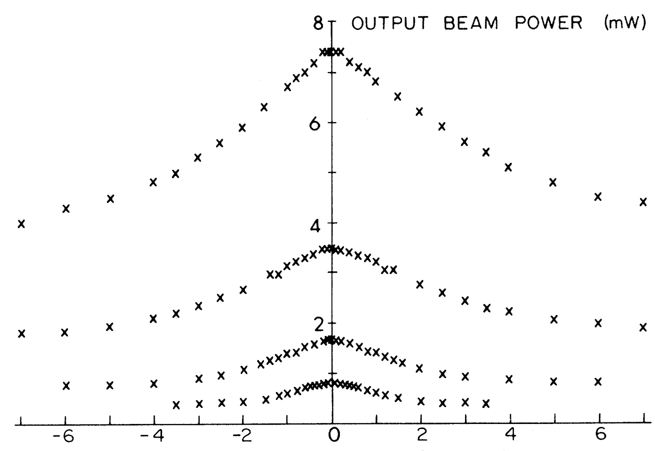

FIG. 4. Energy coupling data: the transmitted power of the amplified beam vs the frequency shift δf between the two beams. The beams were identical to the writing beams in the four-wave-mixing experiments of Fig. 3.

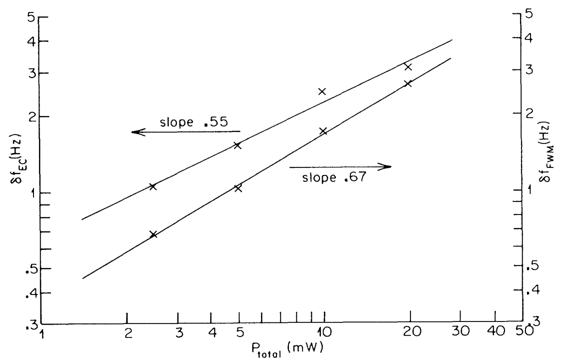

FIG. 5. Frequency shifts vs total incident optical power: Top curve: frequency shift at half maximum of energy coupling; Bottom curve: frequency shift at four-wave-mixing peak. The slopes of these curves yield the intensity dependence of the response rate $1/\tau \sim I^{0.6}$.

In the four-wave-mixing experiments described above, we controlled the powers, frequencies, and directions of the three incident beams, as well as the coupling strength. However, in a self-pumped phase conjugator only the incident beam is provided, and the other beams are generated from self-induced gratings in the conjugator itself.[2] In order to reach a stable state, the gratings increase their diffraction efficiency by translating. Our work here suggests that the wavefront curvatures of the self-generated beams may play a role in determining the sign of the frequency shift. Indeed, it has been experimentally observed that the curvature of the input wave can affect the sign of the frequency shift.[7]

Self-generated beams confined to a ring or linear resonator cavity must satisfy a resonance condition to be stable: The phase of the beam must repeat after one complete trip through the resonator. If the cavity is not exactly an integral number of wavelengths in length, a frequency offset δf occurs spontaneously in the resonating beam.[18,19] Also, in the presence of a uniform dc electric field, a frequency shift occurs which optimizes the phase shift between the intensity fringes and the refractive-index grating.[20] However, we emphasize that neither of the above effects causes the four-wave-mixing enhancement reported here, nor the frequency shifts observed in a self-pumped phase conjugator[2]: The beams are not confined to a closed resonator cavity, and the energy-coupling data in Fig. 4 show that any intrinsic or photovoltaic electric field in the $BaTiO_3$ crystal is negligible.

In conclusion, we have shown that for large coupling strengths, the four-wave-mixing efficiency in photorefractive materials can be increased when a frequency shift between the two writing beams causes the refractive-index grating to translate. The optimum frequency shift has a power-law dependence on the total intensity of the incident optical waves. These results explain the frequency shifts previously observed in self-pumped phase conjugation in a photorefractive $BaTiO_3$ crystal.

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