Observation of Diminished Specular Reflectivity from Phase-Conjugate Mirrors

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We observe that the *specular* reflectivity from the input face of a $BaTiO_3$ phase-conjugate mirror *decreases by over 600%* relative to the standard Fresnel reflectivity value upon the onset of phase conjugation, or wave-front reversal. Reasonable agreement is obtained using a model involving the destructive interference of the Fresnel-reflected beam with a series of phase-conjugate waves generated internal to the crystal. The basic diminishing effect should be universal and hence observable in other classes of self-pumped and externally pumped phase conjugators.

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The field of nonlinear optical phase conjugation[1] has attracted much interest in both applied and fundamental areas of quantum electronics since its inception in the early 1970's. Wave-front reversal has been demonstrated in most states of matter using myriad nonlinear optical mechanisms including stimulated scattering and parametric interactions. Although the wave-front-reversal nature of these interactions has been intensely studied, no study to our knowledge has been undertaken to characterize the *specular* reflection properties of an *isolated* phase-conjugate mirror. Recently, the specular reflection properties of a Fabry-Perot cavity consisting of a dielectric interface and a semi-infinite phase-conjugate mirror have been investigated;[2] the present study, however, if fundamental to a phase-conjugate mirror itself.

In this Letter, we report on the observation of a significant diminishing of the specular reflectivity from the input surface of a phase-conjugate mirror (PCM). In the case of a $BaTiO_3$ PCM, the near-normal specular reflectivity (in air) was observed to decrease from the standard Fresnel value of $\approx 17.8\%$ to $\approx 2.8\%$ upon the onset of the conjugation process. This striking decrease cannot arise simply from an intensity-dependent refractive-index change at the dielectric interface; indeed, this would require that the index decreases from ≈ 2.45 ($BaTiO_3$) to ≈ 1.4. The required nonlinear index would thus have to be orders of magnitude larger than any previously reported, given our operating intensities ($\approx$ W/cm^2).

We deduce that the diminishing effect stems from the destructive interference of a beam undergoing Fresnel reflection at the surface with a *previously unreported beam emerging from the PCM*. The latter beam stems from a sequence of successive conjugation interactions internal to the crystal: A "conventional" volume conjugator which leads to a wave-front-reversed replica, followed by a *previously unreported conjugation process that occurs within one beam diameter of the front surface of the PCM,* and which is mediated via a four-wave mixing[1] interaction. These two internal conjugate mirrors can be shown[3] to be locked in phase relative to each other for all incident angles and wave fronts. The beams generated by this pair of conjugate mirrors combine to interfere destructively with the Fresnel-reflected beam —thereby diminishing its intensity—and, in the process, interfere constructively with the conjugate wave —thereby increasing the externally measured phase-conjugate reflectivity of the conjugator as a whole. The magnitude of the effect depends on the specific nonlinear mechanism(s) internal to the medium, and thus may be material, intensity, and geometry (angle, beam size) dependent. The basic diminishing effect should be universal, and hence observable in other classes of self-pumped and externally pumped PCM's.

We first discuss the basic effect, followed by a description of experimental observations that validate the above conjecture (while ruling out other potential mechanisms), and conclude with a comparison of experimental measurements with model calculations. Various experimental diagnostics lend credence to our model, including the spatial, temporal, polarization, angular, and frequency dependence of the interacting beams,[3] as well as selective optical erasure of the various photorefractive-induced gratings within the crystal.

We assume that all the interacting fields are monochromatic at radian frequency ω_q, with the qth field denoted by

$$\mathbf{E}_q(\mathbf{x}) = \mathbf{A}_q(\mathbf{x}) e^{i[\omega_q t - \mathbf{k}_q\cdot\mathbf{x} + \phi_q(\mathbf{x})]}, \tag{1}$$

where $\mathbf{A}_q(\mathbf{x})$ is real, $\mathbf{k}_q$ is the field wave vector, $\mathbf{x}$ is the propagation direction, and $\phi_q(\mathbf{x})$ is a phase factor depicting the wave front of the field. The basic geometry, along with the two conjugate regions, are sketched in Fig. 1.

Given an input beam, $\mathbf{E}_1$, at frequency ω, the field entering the crystal is $\mathbf{E}_2 = t_F\mathbf{E}_1$, where t_F is the amplitude Fresnel transmission coefficient of the air/crystal interface. The volume phase-conjugate region (PCM_1) generates a conjugate replica of $\mathbf{E}_2$, defined as $\mathbf{E}_3$, and is Stokes shifted in general at frequency $\omega - \delta$:

$$\mathbf{E}_3(\mathbf{x}) = t_F^* r_1 \mathbf{A}_1(\mathbf{x}) e^{i[(\omega-\delta)t + \mathbf{k}_1\cdot\mathbf{x} - \phi_1(\mathbf{x})]}, \tag{2}$$

where r_1 is the complex amplitude reflectivity of PCM_1 which, in our case, is due to self-pumping[4] in $BaTiO_3$. The detailed mechanisms describing PCM_1—including a random overall phase factor associated with the buildup of stimulated scattering processes from statistical noise fluctuations[1]—are of no consequence to realize the di-

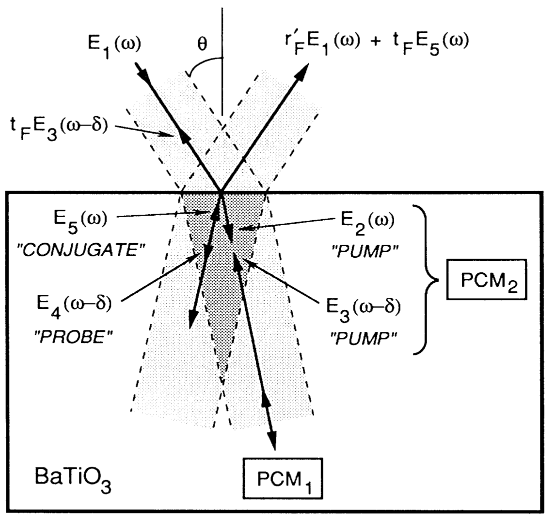

FIG. 1. Schematic diagram of two phased-up conjugation regions that result in a diminished specular reflectivity. PCM_1: Volume conjugator (precise location within crystal not crucial); PCM_2: near-surface conjugator (darker-shaded area).

minishing effect; all that is required is the presence of a conjugate wave within the medium.

The near-surface conjugation region (PCM_2), with amplitude reflectivity r_2 (in magnitude), is mediated by a nearly degenerate four-wave mixing process:[1] The two "pump" beams required for the interaction consist of the incident beam within the crystal ($\mathbf{E}_2$) and its conjugate replica, $\mathbf{E}_3$. These two fields yield a conjugate pair of pump beams, resulting in a phase locking of both PCM's. The "probe" beam incident upon PCM_2, denoted by $\mathbf{E}_4$, is derived from the fraction of the conjugate wave generated by PCM_1 which is internally reflected at the entrance interface into the interaction region of PCM_2, so that $\mathbf{E}_4 = r_F\mathbf{E}_3$, where r_F is the amplitude Fresnel reflection coefficient of the crystal/air interface. As a result of the interaction of the probe wave with PCM_2, a conjugate wave, $\mathbf{E}_5$, at frequency $\omega = \omega + (\omega - \delta) - (\omega - \delta)$ is generated:

$$\mathbf{E}_5(\mathbf{x}) = |t_F r_1 \mathbf{A}_1|^2 t_F r_F^* r_2 \mathbf{A}_1(\mathbf{x}) e^{i[\omega t - \mathbf{k}_1\cdot\mathbf{x} + \phi_1(\mathbf{x})]} . \quad (3)$$

A fraction of this wave, $\mathbf{E}_6 = t_F\mathbf{E}_5$, exits the crystal in the direction of the specularly reflected incident beam, $r_F'\mathbf{E}_1$, where r_F' is the amplitude Fresnel reflection coefficient of the air/crystal interface. These beams, as well as the subsequent reflections, coherently combine, resulting in a total field

$$E_{\text{spec}}(\mathbf{x}) = \{r_F' + |t_F r_1 \mathbf{A}_1|^2 r_2 [t_F^2 r_F^*] + \cdots\} \mathbf{E}_1(\mathbf{x}) . \quad (4)$$

Since the phase factor of the product $t_F^2 r_F^*$ differs by π relative to that of r_F', the internally generated wave *destructively* interferes with the specularly reflected incident wave, yielding the diminishing effect. Note that the exiting wave has the same radian frequency (ω) and

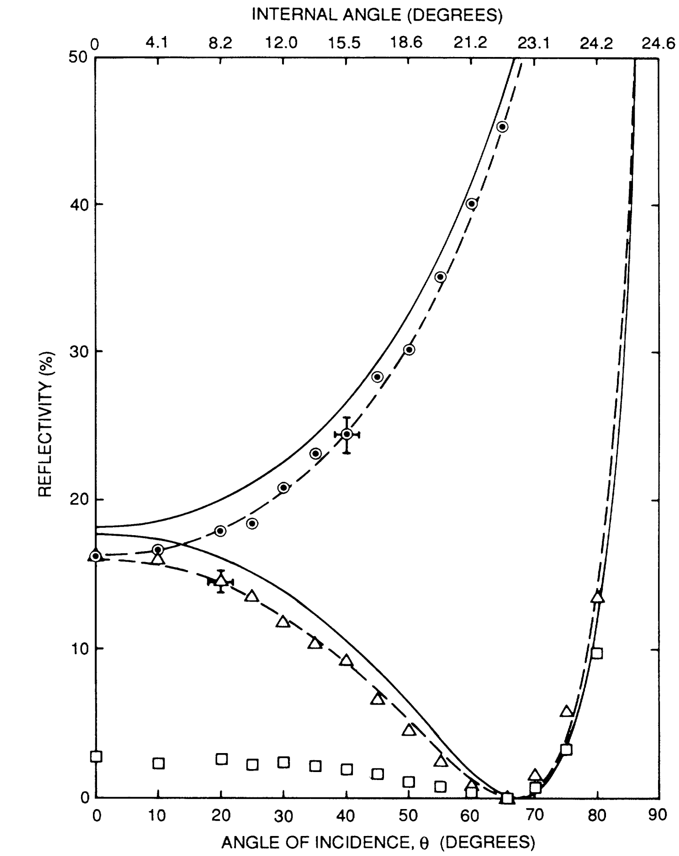

FIG. 2. Specular reflection data for a 45°-cut crystal. s polarization (dots): No conjugation and beam fanning occur for this polarization. p polarization: Stationary crystal (squares) and angularly dithered crystal (triangles); self-pumped conjugation and fanning occur (do not occur) for the stationary (angularly dithered) crystal. Solid curves: Calculated Fresnel reflectivity for an air/$BaTiO_3$ interface using $n_o = 2.488$ and $n_e = 2.423$, after Ref. 6. [Note: Best fit to our data (dashed curves) was obtained using $n_o = 2.354$ and $n_e = 2.310$; the origin of the small discrepancy is not clear, yet does not affect the basic premise of this paper.]

phase front $[\phi_1(\mathbf{x})]$ as the initial specularly reflected wave, $r_F'\mathbf{E}_1$.

The experimental apparatus consists of a single-domain crystal of $BaTiO_3$, with a single-longitudinal-mode cw argon-ion laser ($\lambda = 514.5$ nm, $I \approx 440$ mW/cm^2) as the optical source. For this study, both 0°- and 45°-cut crystals[5] were employed. Detectors are used to monitor the specularly reflected beam, conjugate wave, and the residual on-axis beam transmitted (within a $\approx 1°$ field of view) through the crystal.

The specular reflectivity of the 45°-cut sample as a function of the angle of incidence, θ, for both linear polarization states is shown in Fig. 2. For s polarization, reasonable agreement is obtained with the standard Fresnel reflection coefficient using the accepted values[6] for the refractive index of $BaTiO_3$. For this polarization and crystal orientation, self-pumped conjugation does not occur. Similar results were obtained for both crystal cuts.

For p polarization, a dramatic decrease in the specular reflectivity is seen relative to that calculated using the standard Fresnel relations. At near-normal incidence, the reflectivity is 600% smaller than the Fresnel values for the 45°-cut crystal, and is 30% smaller than the Fresnel value for the 0°-cut sample. Since the diminishing effect is seen to occur for all angles of incidence, the effect is *not* due to a fortuitous succession of internal reflections and subsequent interference with the Fresnel-reflected beam. Thus, a well-defined internal beam (in angle and phase) emanates from the conjugator, resulting in the diminishing effect.

When the crystal is angularly dithered about an axis normal to the plane of incidence at a rate (≈ 100 Hz) faster than the characteristic grating buildup time, and through an excursion ($\Delta\theta \approx 0.5°$) in excess of the Bragg acceptance angle, the conjugate wave as well as beam fanning[1] both vanish. Under these conditions, the angular dependence of the specular reflectivity for the p-polarization state is seen to agree reasonably well with the Fresnel-calculated values, as plotted in Fig. 2. Thus, *the diminishing effect has its origins in the nonlinearly induced gratings,* and is not due to an anomalously large nonlinear index or other scattering effect.

The temporal evolution of the specularly reflected beam, conjugate wave, and on-axis crystal throughput is shown in Fig. 3. Since the onset of beam fanning (which temporally precedes the conjugation process) deflects most of the beam off axis, the output of the on-axis detector is seen to decrease in time. On the other hand, the conjugate-wave buildup is temporally correlated with the decrease in the specularly reflected beam. Thus, *the diminishing effect is intimately related to the presence of the conjugate wave within the medium,* and is not related (at least directly) to beam fanning.

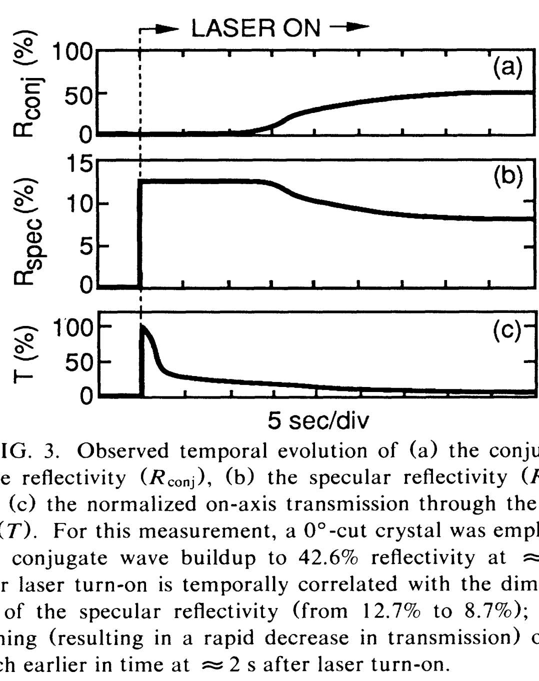

FIG. 3. Observed temporal evolution of (a) the conjugate-wave reflectivity (R_{conj}), (b) the specular reflectivity (R_{spec}), and (c) the normalized on-axis transmission through the crystal (T). For this measurement, a 0°-cut crystal was employed. The conjugate wave buildup to 42.6% reflectivity at ≈ 15 s after laser turn-on is temporally correlated with the diminishing of the specular reflectivity (from 12.7% to 8.7%); beam fanning (resulting in a rapid decrease in transmission) occurs much earlier in time at ≈ 2 s after laser turn-on.

Finally, an incoherent beam with an intensity 10 times that of the probe beam was employed to selectively erase[7] small (≈ 0.2 mm diam) spatial regions of the optically induced gratings in the crystal; illumination was normal to the plane of incidence. In one case, the central portion of the crystal (PCM_1) was illuminated by the erase beam, resulting simultaneously in a restoration of the specularly reflected beam to its Fresnel-calculated value and a complete vanishing of the conjugate beam. When the erase beam illuminated the crystal near the input surface (PCM_2), the specularly reflected beam was again restored to its Fresnel-calculated value; however, most of the conjugate-beam flux persisted (a 40% decrease was observed in the conjugate reflectivity, attributed to optical scattering of the erase beam within the crystal). Hence, we conclude that *two distinct grating regions exist within the crystal:* one which is required for the volume conjugator, and a second near-surface conjugator that contributes to the cancellation of the specular component.

To analyze our system, we employed a generalized treatment of the diminished specular reflectivity model[2] that includes *two* distinct, yet phase-locked PCM's, obtained by summing the terms in Eq. (4). The model predicts a diminished specular (power) reflectivity (R_{spec}), given the Fresnel reflectivity (R_F), and that of the two internal PCM's ($R_{1,2}$):

$$R_{spec} = R_F\{[1-(R_1R_2)^{0.5}]/[1-R_F(R_1R_2)^{0.5}]\}^2, \qquad (5)$$

where $R_i = |r_i|^2$. The model also predicts an enhanced overall conjugate reflectivity, R_{conj}:

$$R_{conj} = R_1(1-R_F)^2/[1-R_F(R_1R_2)^{0.5}]^2. \qquad (6)$$

Given the externally measured quantities, R_{spec} and R_{conj}, Eqs. (5) and (6) can be solved for the reflectivities of the internal PCM's. From R_1 and typical interaction parameters of the near-surface conjugator—the two-wave gain-length product[8] and the amplitude ratios of the three interacting beams ($\mathbf{E}_2$, $\mathbf{E}_3$, and $\mathbf{E}_4$)—we can calculate[3] its reflectivity, R_2', using a depleted pump analysis.[9] Using the values of R_2' and R_1 in Eq. (5), we arrive at a predicted value of the diminished reflectivity (R_{spec}'), which we compare with our measurements. Results for an internal angle of incidence of 10° are tabulated in Table I. The difference in the magnitude of the diminishing effect for the two crystal cuts arises primarily from their unequal two-wave gain-length products: 0.376 and 1.88, for the 0°- and 45°-cut crystals, respectively. The close quantitative agreement of theory and experiment is fortuitous, given the variation of material parameters and the precise beam overlap geometry.

TABLE I. Calculated and measured specular and conjugate reflectivities for both 0°- and 45°-cut crystals using p-polarized light at an internal (external) angle of 10° ($\approx 24.2°$).

	Specular reflection			Conjugate Reflection		
Crystal cut	Calculated Fresnel refl. R_F (%)	Observed specular refl. R_{spec} (%)	Calculated specular refl. R'_{spec} (%)	Meas. R(PCM) R_{conj} (%)	Calc. $R(PCM_1)$ R_1 (%)	Calc. $R(PCM_2)$ R'_2 (%)
0°	12.7	8.7	9.9	42.6	53.7	3.2
45°	13.2	2.4	2.1	61.4	68.4	59.3

Nonetheless, the observed diminishing effect for the two crystal cuts, coupled with the series of parameter studies, give us confidence that the physical mechanism is well characterized.

In conclusion, we have observed a significant diminishing of the specular reflectivity from self-pumped phase-conjugate mirrors. We have since observed similar diminishing effects in self-pumped conjugators using the "external loop" configuration[10] in $BaTiO_3$ and in $KNbO_3$ (Ref. 11), as well as in $BaTiO_3$ using the standard externally pumped four-wave mixing geometry.[1] The basic diminishing effect should be universal and hence observable in other classes of PCM's, including resonantly enhanced nonlinear media such as sodium vapor,[12] and in stimulated Brillouin scattering PCM's, in which case the near-surface conjugate region may be mediated via a Brillouin-enhanced four-wave mixing process.[13] A practical consequence of the diminishing effect is that unless taken into account, one can inadvertently overestimate the internal volume phase-conjugate reflectivity and hence overestimate the nonlinear susceptibility, as well as underestimate the linear refractive index of the medium. The diminishing effect is expected to be most pronounced in materials possessing large linear refractive indices and nonlinear susceptibilities, such as semiconductors and electro-optic oxides.

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