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TITLE: HIGH EFFICIENCY PHASE-CONJUGATE REFLECTION
IN GERMANIUM AND IN INVERTED CO₂

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HIGH EFFICIENCY PHASE-CONJUGATE REFLECTION
IN GERMANIUM AND IN INVERTED CO₂

by

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Abstract

We report the first observation of phase-conjugate reflection at 10.6 μm . We used a novel intracavity technique which is of general utility for any oscillating laser system. In one experimental arrangement, we placed germanium in a TEA CO₂ laser cavity to utilize the intracavity counterpropagating (strong) waves for degenerate ℓ -wave mixing. Reflectivities as high as 20% were obtained. In a second experimental arrangement, phase-conjugate reflection and amplification with an effective gain exceeding unity was obtained by redirecting the output of a TEA CO₂ laser oscillator into the active gain medium. The intense counter-propagating waves within the laser medium coupled with the saturated gain medium to provide the nonlinearity in a process analogous to degenerate four-wave-mixing.

Introduction

We have generated a phase-conjugate, 10.6 μm , reflection from the phase grating established with counter-propagating waves in both germanium samples located within an oscillating TEA double-discharge CO₂ laser cavity and in the CO₂ gain medium itself. The work in germanium is the first demonstration of nonlinear phase conjugation in the infrared, and, more generally, of intracavity degenerate 4-wave mixing. The work within the CO₂ gain medium is the first demonstration of infrared phase-conjugation in an inverted medium. These intracavity techniques have general applicability for any laser system.

The concept of phase-conjugate reflection was first discussed by Zel'dovich and coworkers.¹ In such a process, a nonlinear interaction (such as stimulated Raman scattering, stimulated Brillouin scattering, or degenerate four-wave-mixing) gives rise to a "reflected" wave which is the amplitude complex conjugate of the incident wave. The signature of the process is the fact that a phase-conjugate reflection exactly retraces the optical path of an incident wave, regardless of any phase distorters which may be in the path of the incident wave.

To understand the difference between conventional mirror reflections and phase-conjugate reflections, let us examine Figure 1. The left-hand side depicts a conventional mirror in which the incoming and outgoing rays are related by inversion of only the component of the K-vector normal to the mirror surface. It is this relationship which gives us the standard image - transformation properties for shiny optical surfaces. The right-hand side of Fig. 1 depicts the action of a phase-conjugate reflector; note that rays are bent back upon themselves. In such a reflection the vector quality \vec{K} is changed in sign, and the device's image transformation properties are far different. A person looking into a conventional mirror would see his face, whereas a person looking into a phase-conjugate mirror would only see the pupil of his eye. This is because any light emanating from his cheek, for instance, would, upon conjugation, be returned to his cheek and would therefore miss his eye.

Figure 2 shows how conjugate optics can improve the double-pass operation of laser amplifiers. Consider a high quality optical beam entering from the left. As it traverses the optical system it becomes distorted because of imperfections, improper surfaces, inhomogeneities in the gain medium, etc. If the distorted beam is reflected by a phase-conjugator, it is redirected in such a way that each distorted ray returns to its corresponding aberrating spot and the beam emerges with excellent optical quality. This would mean that large optical systems could be arranged so that there are many portions whose imperfect optical quality would not impair the passage of a diffraction-limited beam, and this could result in large savings high-power laser designs.

Among the many potential applications of such a phase-conjugator, the application to laser-fusion is perhaps the most exciting. Figure 3 shows how conjugate optics could modify approaches to the laser-fusion program. In this arrangement, the fusion target is diffusely illuminated. The scattered light could travel backwards through the amplifier chain, could be reflected by a conjugating 4-wave mixing medium, would return through the imperfect optical elements, and an amplified pulse would impinge directly on the target, obviating the need for ultra-precise target alignment. Such a phase-conjugating mirror will undoubtedly find other applications, such as providing stable laser operation even in a gain medium experiencing time-varying inhomogeneities, and possibly diagnosing laser-plasma interactions.

The subject of phase-conjugate reflection via nonlinear optical effects has received much recent attention in the literature. Stimulated Brillouin scattering,^{1,2} stimulated Raman scattering,³ three-wave

CONVENTIONAL MIRROR PHASE-CONJUGATE MIRROR

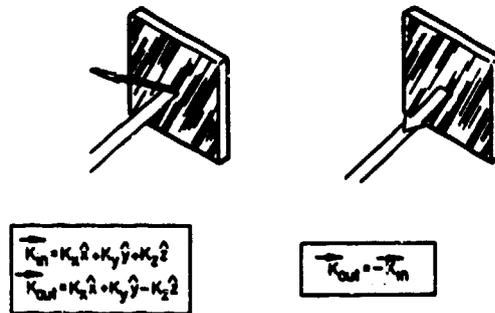


Fig. 1 Comparison of reflections from a conventional mirror and a phase-conjugate mirror.

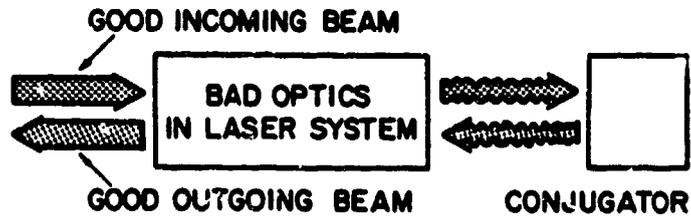


Fig. 2 An application of phase conjugate reflection to the restoration of optical quality in double-passing an imperfect optical system.

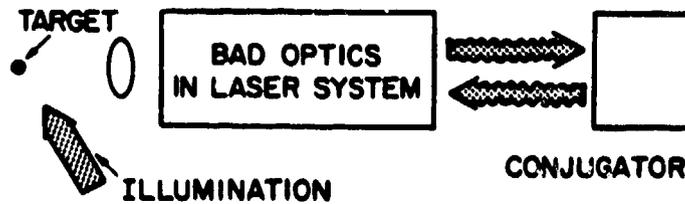


Fig. 3 A potential application of phase-conjugate optics to laser fusion. The diffusely illuminated target scatters some light through the laser amplifier system; upon conjugate reflection and subsequent reamplification, the bright pulse strikes the target.

mixing,⁴ and degenerate four-wave mixing⁵⁻⁷ have all been utilized to generate a conjugate backward reflection, and a review article by Yariv⁷ has recently appeared.

How Four-Wave Mixing Generates Phase Conjugation

For the simplest example of this effect, consider a material which the index of refraction depends nonlinearly upon intensity. Such a material would, for example, exhibit strong self-focusing or strong self-defocusing (depending upon whether the refractive index increases or decreases with intensity). The nonlinear response can be described as an index of the form $n = n_0 + n_2 \langle E^2 \rangle$, or as a polarizability $\chi = \chi^{(0)} + \chi^{(3)} E^2$. The coefficients are related through $n_2 = 2\pi \chi^{(3)} / n_0$.

Consider waves E_1 , E_2 , and E_3 (all of the same frequency) propagating through this nonlinear material as shown in Fig. 4. E_1 and E_2 are arranged to be precisely counterpropagating plane waves, and they are further assumed to both be far stronger than E_3 . In the simplified theory, the pair of oppositely propagating (pump) waves (at the same frequency ω) in a material exhibiting a nonlinear index of refraction, provides the conditions for which a third (probe) wave (E_3), also at frequency ω , incident on the material (from any direction) would result in a fourth wave being emitted from the sample precisely retracing the k -vector of the third wave. Under the above conditions, phase matching is guaranteed (even in birefringent materials) independent of the angle between k_3 and the pump waves k_1 and k_2 . For E_3 , $E_4 \ll E_1, E_2$, in the absorption-free case $E_4 = E_3^* (\tan \kappa / \kappa) \lambda$, where κ is the strength of the nonlinear mixing process ($\kappa = (2\pi/\lambda_0) n_2 E_1 E_2$) and λ is the geometric interaction length. Note also that the wave E_4 is proportional to the complex-conjugate of E_3 ; this is the essential feature of the phase conjugation effect. Note that, although a nonlinear phenomenon is utilized, the effect is linear in the field one wishes to conjugate. This is why this technique is far more attractive than, say, stimulated Brillouin backscattering. Because our effect is linear, a superposition of E_3 's will generate a corresponding superposition of E_4 's, so one can readily see the generalization of the effect; any complex field is time-reversed in such an arrangement.

It is valuable to consider how the interference gives a conjugated E_4 wave. Consider first the interaction of the weak wave E_3 and one strong wave E_1 . The index change contains the term $(E_1 + E_3)^2$, which contains a cross-term corresponding to a phase-grating perfectly phase-matched so that the other strong wave, E_2 , is Bragg-scattered into the E_4 wave. Concurrently E_2 and E_3 interfere to form a phase grating which scatters E_1 into the E_4 wave. This process is analogous to volume holography in which the writing and reading are done simultaneously.

Experimental Results in Germanium

Numerous demonstrations of intracavity, phase-conjugate reflection via degenerate 4-wave mixing in the visible have appeared recently.^{6,7} These experiments had utilized beams from frequency-doubled YAG lasers, from dye lasers, from ruby lasers, and from cw argon-ion lasers; 4-wave mixing media have included liquid CS_2 , resonant, near-resonant, and two-photon resonant absorbing sodium vapor, and rubi crystal, yet prior to our work, no experiments had been done at $10.6 \mu m$. Demonstrating this effect in the infrared was made difficult because: 1) The effect has an ω^2 dependence, and, thus, is reduced a factor of 400 from the corresponding visible effect. 2) Very strong pump waves are needed, and 3) the pump waves must be perfectly collimated and precisely retrodirected.

We recognized that one could reduce the constraints upon the pumpwaves by requiring that they merely be complex conjugates of each other. This means that if E_1 is diverging, E_2 must be converging, and vice versa. Since the two counterpropagating waves inside an oscillating laser cavity are already complex-conjugates of each other, if the nonlinear material is placed within the cavity of an oscillating stable-mode laser, the material is guaranteed to be a conjugator for light of the laser frequency. As an added bonus, the circulating power within a laser can be much higher than the output power.

In our first experiments⁸ we used a hybrid low-pressure/high pressure TEA CO_2 laser⁹ operating on the P(20) line with an output intensity of $2 MW/cm^2$ in a temporally smooth pulse of 40 nsec FWHM duration. The output beam diameter was 0.4 cm. As depicted in Fig. 5, the slightly wedged, intrinsic, polycrystalline, germanium flat output mirror (R=95%) was reversed so that both the antireflection-coated surface and the 0.5-cm-thick germanium substrate were internal to the optical cavity. The substrate of the germanium output coupler itself therefore served as the nonlinear medium. This simple intracavity technique eliminated the difficulty of precisely aligning a pair of counterpropagating beams in the nonlinear sample. Germanium was chosen for our experiment because it was readily available and has¹⁰ an exceptionally large third-order susceptibility ($\chi^{(3)} = 1.5 \times 10^{-10}$ esu). The laser output was then routed by mirrors through a 50% ZnSe beamsplitter to impinge upon the "active" region of the germanium output coupler. A ZnSe lens ($f = 14$ cm) was placed at a distance $1.5f$ in front of the germanium; this lens improved the overlap by partially concentrating the beam to be conjugated (probe beam) on the active region. The phase-conjugated beam was then retrodirected by the beamsplitter along the remainder of a 6-meter-long path and then through an adjustable aperture into a high-speed HgCdTe detector for diagnostics. A removable, flat, 100% reflective mirror could be placed in front of the lens and accurately aligned to provide a reference reflection against which the back-scattered signals were compared.

The peak power of the phase-conjugated beam passing through the 6-mm-diameter aperture corresponded to a reflectivity of 2%, in accord with theoretical predictions, whereas the simple scattering through the same



Fig. 4 The basic geometry of four-wave mixing. The arrows depict the orientations of the K-vectors.

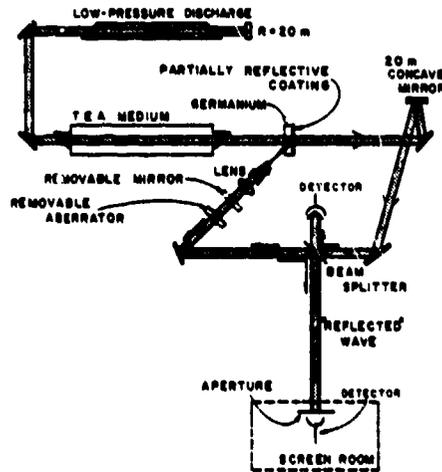


Fig. 5 Experimental setup for obtaining phase conjugate reflection in the germanium substrate of the inverted output coupler.

aperture (measured with only the low-power, low-pressure section of the hybrid laser operating) was less than 2×10^{-5} of the power measured with the reference mirror in place. Appropriate beam-blocking tests were performed to distinguish the signal from other sources of scattering. The canonical test for phase conjugation, the placement of a severely aberrating element in the beam line, had little effect on the reflected signal after re-passing the aberrating element, whereas the same aberrator in front of the reference mirror reduced the reflected signal by more than 99%.

For a somewhat qualitative but exceedingly demonstrative illustration of the effect, an infrared-sensitive, pyroelectric vidicon replaced the detector and aperture in the screen room. Photographs of the video monitor are shown in Fig. 6. To obtain sufficient sensitivity, we placed a 1"-diameter collecting lens in front of the vidicon which focused the $10.6 \mu\text{m}$ radiation to a plane well beyond the vidicon's sensitive surface. As can be seen in Fig. 6b, the phase conjugate signal is quite compact and in Fig. 6a is hardly modified by double-passing the aberrator 6 meters away, whereas the aberrator has a very deleterious effect upon the signal returned by the reference mirror (Fig. 6c). The aberration in Fig. 6c are probably more severe than the photograph indicates because of truncation by the finite aperture of the collecting lens.

The temporal behavior of the laser and phase-conjugate signals were recorded on a fast oscilloscope. Typical traces are shown in Fig. 7. For comparison, both cases depicted have the same vertical and horizontal deflections. To avoid problems associated with possible detector nonlinearities, calibrated CaF_2 attenuators were placed in front of the detector to keep the electrical signals roughly equal. Fig. 7a shows the reference-mirror reflected pulse with a 100-fold attenuator in front of the detector; the FWHM duration is approximately 50 nsec. Fig. 7b shows the conjugate-reflected pulse with no attenuation in front of the detector; the shortening of the pulse $\sim 1/\sqrt{3}$ (as simple theory predicts) is clearly evident as is the time 2% reflectivity of the nonlinear effect.

In later experiments, we used thicker germanium samples. A 1" thick 95% R coated end-piece gave a conjugate reflectivity of 10%, whereas a 15-cm-long single-crystal boule placed in the center of the cavity gave 20% conjugate reflection efficiency. The effect did not scale simply as λ^2 , because as λ of the nonlinear sample was increased, the circulating power within the cavity decreased. We attribute this to the intensity-dependent formation of an electron-hole plasma¹¹ in the germanium.

Conjugate Reflection In Inverted CO_2

In the second set of experiments, we chose to obtain conjugation utilizing the nonlinear properties of the resonance associated with the CO_2 gain medium itself, and, again, our intracavity technique used the counterpropagating waves already present in the oscillating laser cavity. As an added bonus when working with the nonlinearity of a partially saturated gain medium, if the probe beam is directed through "unused" gain volume on its way to (and, of course, from) the interaction region, then the phase-conjugate signal is automatically amplified.

The nonlinear process utilized in the CO_2 gain resonance is not merely the four-wave mixing process; because of saturation the effect is the coherent superposition of four-wave, six-wave, eight-wave, ... processes. Because of the ambiguity in the number of waves involved, we prefer to identify this mixing process as resonant light-by-light scattering. The theory of phase-conjugate resonant light-by-light scattering was first developed by Abrams and Lind.¹² These authors utilized the well-known nonlinear susceptibility of a homogeneously-broadened two-level absorber (or amplifier) which, on resonance, sets up an amplitude grating which couples the standing field and the probe field to generate the conjugate wave. As should be expected, the maximum scattering efficiency is attained when the strong counterpropagating standing waves only partially saturate the transition. This is because the depth of the amplitude grating becomes reduced as the strong waves exceed the saturation flux, and this is related to the reason that the effect is not merely a third order (four-wave) process.

In this experimental setup, our conventional hybrid high-pressure/low-pressure TEA laser is operated with a 10 m radius of curvature 35% reflecting mirror and a flat 10% reflecting mirror. These unusually low reflectivities were chosen to reduce the circulating power within the cavity. The laser operated on the P(20) line of the $10 \mu\text{m}$ branch, and the pulse emitted from the curved output coupler was approximately 150 nsec (FWHM) in duration and had an energy of approximately 100 mJ. This corresponded to a circulating intensity of approximately $1.5 \text{ MW}/\text{cm}^2$ inside the cavity. The output of the laser was attenuated, passed through a beam splitter and redirected back through the TEA laser gain medium. Phase conjugate reflectivity was diagnosed using an infrared vidicon or a high speed SAT detector on the return signal reflection from the beam-splitter. The phase conjugate nature of the reflected light was confirmed by studying the spatial characteristics of the beam in the presence of an aberrator and in the presence of a cross-hair image placed on the beam prior to the beamsplitter. In both cases the initial incident probe beam image is preserved in the reflected image. In a single pass configuration where the probe beam overlapped the lasing region of the gain medium only in the last 10 cm of the 91 cm discharge length, a 1/2% reflectivity was observed; reflectivity being defined as the peak intensity of the phase conjugate beam divided by the peak intensity of the incident probe beam. In a double and triple pass arrangement where again only the last 10 cm of the final pass through the gain medium overlapped the lasing region reflectivities of 35% and 250% were observed, respectively. The enhanced reflectivity in these cases is due entirely to the amplification of the outgoing probe and incoming phase conjugate signal in the en route to and from the conjugation region. The actual phase-conjugate reflectivity in the 10 cm overlap region is in all cases less than 10^{-2} , in accord with the theory of Abrams and Lind¹² for the field parameters and gain medium parameters involved.

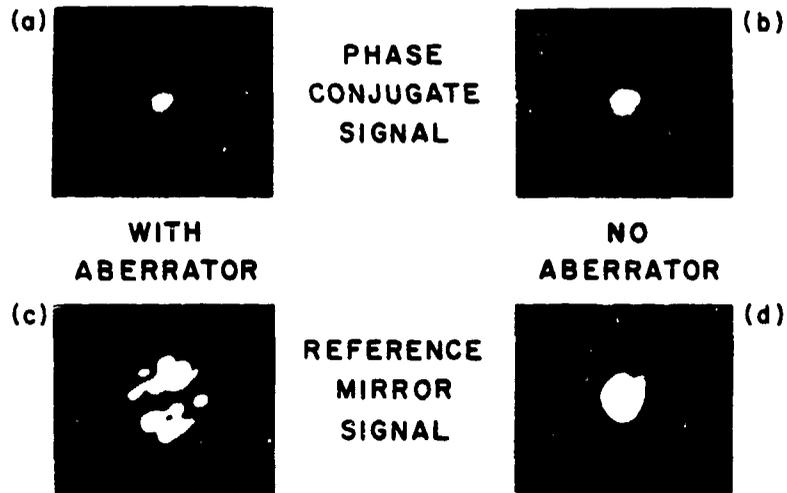


Fig. 6 Video displays of spatial patterns. Note that insertion of the aberrator has virtually no effect upon the quality of the phase conjugate-reflected beam, but that it badly distorts the mirror-reflected beam.

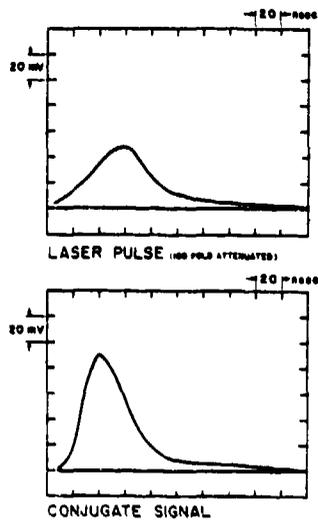


Fig. 7 Temporal record of laser pulse and conjugate signal for the germanium experiment. The shortening of the conjugate signal is in accord with simple theory.

Concluding Remarks

We have demonstrated that a very simple intracavity arrangement is useful in obtaining phase-conjugate reflections. The technique is readily applicable to any nonlinear material placed in an oscillating laser cavity; if the laser operates, the material is guaranteed to be a conjugate reflector for light of that frequency.

Using this technique, we have obtained the first demonstration of phase-conjugate reflection in the infrared using both germanium and the CO₂ gain medium itself.

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References

1. B. Ya. Zel'dovich, V. I. Popovichev, V. V. Ragul'skii, and F. S. Faizullov, Zh. Eksp. Teor. Fiz. Pis'ma Red. 15, 160 (1972) [Sov. Phys. JETP 15, 109 (1972)]; O. Yu. Nosach, V. I. Popovichev, V. V. Ragul'skii, and F. S. Faizullov, Zh. Eksp. Teor. Fiz. Pis'ma Red. 16, 617 (1972) [Sov. Phys. JETP 16, 435 (1972)].
2. V. G. Sidorovich, Zh. Tekh. Fiz. 46, 2168 (1976) [Sov. Phys. Tech. Phys. 21, 1270 (1976)]; I. M. Bel'dyugin, M. G. Galushkin, E. M. Zemskov, and V. I. Mandrosov, Kvant. Elektron. 3, 2467 (1976) [Sov. J. Quantum Electron. 6, 1349 (1976)]; G. G. Kochemasov and V. D. Nikolaev, Kvant. Elektron. 4, 115 (1977) [Sov. J. Quantum Electron. 7, 60 (1977)]; V. Wang and C. R. Giuliano, Opt. Lett. 2, 4 (1978); V. G. Balenko, B. G. Gerasimov, V. M. Podgaetskii, and L. K. Slivka, Kvant. Elektron. 4, 933 (1977) [Sov. J. Quantum Electron. 4, 530 (1977)].
3. B. Ya. Zel'dovich, N. A. Mel'nikov, N. F. Pilipetskii, and V. V. Ragul'skii, Pis'ma Zh. Eksp. Teor. Fiz. 25, 44 (1977) [JETP Lett. 25, 36 (1977)]; N. F. Pilipetskii, V. V. Ragul'skii, V. V. Shkounov, and B. Ya. Zel'dovich, presented at 1977 Vavilov Conference, Novosibirsk, USSR (1977) (unpublished).
4. A. Yariv, J. Opt. Soc. Am. 66, 301 (1976); P. V. Avizonis, F. A. Hopf, W. D. Bamberger, S. F. Jacobs, A. Tomita, and K. H. Womak, Appl. Phys. Lett. 31, 435 (1977).
5. R. W. Hellwarth, J. Opt. Soc. Am. 67, 1 (1977); A. Yariv and D. M. Pepper, Opt. Lett. 1, 16 (1977).
6. D. Bloom and G. C. Bjorkland, Appl. Phys. Lett. 31, 592 (1977); P. F. Liao, N. P. Economou, and R. R. Freeman, Phys. Rev. Lett. 39, 1473 (1977); S. H. Jensen and R. W. Hellwarth, Appl. Phys. Lett. 32, 166 (1978); D. M. Bloom, P. F. Liao, and N. P. Economou, Opt. Lett. 2, 58 (1978); P. F. Liao and D. M. Bloom, Opt. Lett. 3, 4 (1978); P. F. Liao, D. M. Bloom, and N. P. Economou, Appl. Phys. Lett. 32, 813 (1978); D. M. Pepper, D. Fekete, and A. Yariv, Appl. Phys. Lett. 33, 41 (1978); D. Grischkowsky, N. S. Shiren, and R. J. Bennett, Appl. Phys. Lett., 33, 805 (1978).
7. A. Yariv, IEEE J. Quantum Electron. QE-14, 650 (1978).
8. E. E. Bergmann, I. J. Bigio, B. J. Feldman, and R. A. Fisher, Opt. Lett. 3, 82 (1978).
9. A. Goldhaiekar, N. R. Heckenberg, and E. Holzhauser, Phys. Lett. 46A, 229 (1973).
10. J. J. Wynne, Phys. Rev. 178, 1295 (1969).
11. C. R. Phipps, Jr., Private communication.
12. R. L. Abrams and R. C. Lind, Opt. Lett. 2, 94 (1976), and Erratum, Opt. Lett. 3, 205 (1978).