mogeneously distributed stresses that would induce splitting of the $J=1$ level.

[7] T_1 in solid H_2 with high concentrations of ortho was first calculated by T. Moriya and K. Motizuki, Progr. Theoret. Phys. (Kyoto) 18, 183 (1957). The calculation has been repeated recently by A. B. Harris and E. Hunt, Phys. Rev. Letters 16, 845 (1966).

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OBSERVATION OF DEGENERATE STIMULATED FOUR-PHOTON INTERACTION AND FOUR-WAVE PARAMETRIC AMPLIFICATION

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We report in this Letter the observation of stimulated four-photon or light-by-light scattering for the case where the coupling is due to the molecular-orientation Kerr effect. In this process two forward-going photons in an intense beam of light scatter to produce two photons traveling at small angles $+\theta$ and $-\theta$, respectively.[1] If photons are already present in the $+\theta$ and/or $-\theta$ beams, this stimulates the process. In contrast to the stimulated Raman case, coupling of these weak waves produces gain.[2] One type of four-photon or four-wave mixing experiment was performed by Maker and Terhune involving $\chi^{(3)}(\omega+\Delta\omega, \omega-\Delta\omega, \omega, \omega)$ where the beams are all collinear and $\Delta\omega \neq 0$.[3] In our experiment, weak-wave retardation allows phase matching and hence strong exponential gain even for the degenerate case when all the waves have the same frequency and are not collinear.[4] Furthermore, this experiment isolates the fundamental process causing beam trapping, namely, the amplification of nonforward directed Fourier components of the beam.

A Q-switched ruby-laser beam with a power of 150-300 MW and a divergence of 6 mrad was focused by a lens of 1-m focal length into a short liquid cell. A weak beam in the $+\theta$ direction was obtained from the forward laser beam by a beam splitter and mirror arrangement (as shown in Fig. 1). Both beams were then focused and deflected by the same lens so as to cross within a nitrobenzene cell. The $+\theta$ beam is amplified in the process of stimulated light-by-light scattering and, in addition, a beam in the $-\theta$ direction is generated. The appearance of this $-\theta$ beam is the signature of this effect. For the purpose of alignment, a large-diameter He-Ne gas-laser beam was arranged to coincide with the ruby beam. This was used in conjunction with a focusing alignment telescope which assured that the two beams

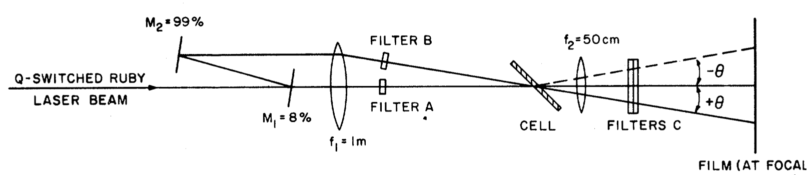

FIG. 1. The experimental arrangement. Q switching was accomplished by a combination of a rotating prism and a saturable dye cell.

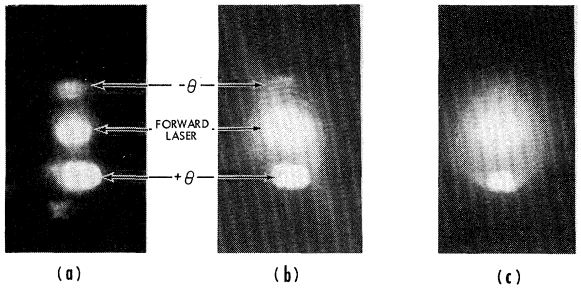

FIG. 2. (a) Typical data for a 3-mm cell length where no self-focusing occurs. Due to overexposure, the actual intensity ratios are not faithfully reproduced. (b) Simultaneous (1.5-cm spacer- Fabry-Perot interferometer analysis of all three beams from a 3-mm cell. (c) Typical data for a 7.5-cm cell length illustrating the nearly symmetrical redistribution of the $+\theta$ and $-\theta$ beams observed when self-trapping is present.

crossed in the focal region and within the cell. A signal was observed for a 3-mm cell length of nitrobenzene and is shown in Fig. 2(a). When the forward laser beam was blocked by a piece of metal, located at the position of filter A, or the $+\theta$ beam was similarly blocked at the position of filter B, the $-\theta$ beam disappeared. This eliminated the possibility of the $-\theta$ beam being a spurious reflection. Also, a simultaneous Fabry-Perot interferometer analysis of all three beams [Fig. 2(b)] showed that each beam contained the same frequency components to within 200 MHz, the laser linewidth. Hence the $-\theta$ beam could not have arisen from stimulated inelastic light scattering, such as backward Brillouin or Rayleigh-wing scattering. An interference filter at the ruby-laser frequency along with Corning 2-64 and 4-97 filters (Filters C in Fig. 1) and Polaroid type-57 film eliminate Raman scattering as the cause of this effect. Also, the threshold for forward Brillouin scattering would be much too high for this configuration, especially since the laser pulse duration was only 14 nsec.

Figure 2(a) was taken with 240 ± 10 MW of forward laser power which was focused inside the Brewster-angle cell to an experimentally measured area of 0.20 ± 0.06 cm^2. The incident power density including optical losses was 900 ± 300 MW/cm^2, assuming a spatially uniform output power. This corresponds to a $\theta_{opt} = (\epsilon_2 |E_0|^2/2\epsilon_0)^{1/2} = 7.2 \pm 1.3$ mrad, where $\epsilon_2 = 5 \times 10^{-11}$ esu for nitrobenzene.[5] Experimentally the axis of the $+\theta$ beam was set at $\theta_{exp} = 8.3 \pm 0.8$ mrad. When θ_{exp} was increased or decreased from this value, the $-\theta$ beam intensity went down. The presence of the two spots in the $-\theta$ beam simply reproduced the forward-beam profile which also contained two spots (not visible because of the saturation of the film).

Consideration of the normal-mode problem with the boundary conditions that $I_{-\theta}(0) = 0$ and $I_{+\theta}(0) \neq 0$ gives[6] at θ_{opt}

$$I_{-\theta}(L) = I_{+\theta}(0)(\sinh\tfrac{1}{2}gL)^2$$

and

$$I_{+\theta}(L) = I_{+\theta}(0)(\cosh\tfrac{1}{2}gL)^2,$$

where L is the cell length and where, for the results shown in Fig. 2(a), the $+\theta$ beam was initially 7 % of forward laser intensity. When the input $+\theta$ beam intensity is decreased by a factor of 10, the $-\theta$ intensity observed is decreased by approximately a factor of 10, in agreement with the above equations. The calculated gain for the above conditions is $g = 2\pi n_0 \theta_{opt}{}^2/\lambda_0 = 7.4 \pm 2.6$ cm^{-1} or $gL = 2.7 \pm 1.0$, since $\theta_{exp} \approx \theta_{opt}$, giving $[I_{-\theta}(L)/I_{+\theta}(L)]_{theory} \cong 0.8$. However, experimentally $I_{-\theta}(L) \approx I_{+\theta}(L)/10$ for the total intensities involved corresponding to $gL = 0.7$. There are several obvious explanations for this being smaller than the theoretical value. First the $+\theta$ beam is not a plane wave, so that only a small fraction of the $+\theta$ beam power is at the correct angle. Second, it is possible that the theory of Chiao, Kelly, and Garmire[4] should be modified to take into account the angular spread of the forward laser beam which will again reduce the gain. Finally, when $I_{+\theta}$ becomes comparable with I_{laser}, the gain may be still smaller.

It is expected that light-by-light scattering develops eventually into a form of self-focusing. Therefore, cell lengths greater than the self-focusing distance for these power densities may be expected to yield results differing from the above. For example, some fraction of the $+\theta$ and $-\theta$ beams may become trapped within the resulting wave guide. If such a process occurs, this may affect the character of the filaments and also cause a nearly symmetrical redistribution of the trapped portions of the $+\theta$ and $-\theta$ beams. This is an explanation of the observation shown in Fig. 2(c) which was taken with a 7.5-cm cell of nitrobenzene under otherwise similar conditions. The observed intensity distribution lost its diffuse background when the $+\theta$ beam was blocked.

By varying the cell length, the pattern shown in Fig. 2(a) made a smooth transition to that of Fig. 2(c).

When the Raman effect was photographed (using a Corning 7-69 as filters C), several interesting things were observed. No Raman light in the forward direction was observed for the 3-mm cell, but it was strongly present for the 7.5-cm cell. For the longer cells, if the $+\theta$ beam was varied from zero to 7% of the forward laser power, the Raman power changed two orders of magnitude. However, when the forward laser power was increased by the same amount, no such marked increase in the Raman effect was observed. Since in this liquid the Raman effect is intimately related to self-focusing,[7] possible interpretations are that the presence of the $+\theta$ beam changes the focusing characteristics so that higher fields exist at the focus, or that focusing occurs closer to the cell entrance window which then increases the effective gain length.

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[1]The kinematics of photon-echo experiments [I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. 141, 391 (1966)] is identical with that described here, but the lack of time delay and the transparency of the nonlinear materials used rule this out for the case discussed.

[2]Although the present process arises from $\chi^{(3)}$, as does the stimulated Raman effect, a further point of contrast is that stimulated light-by-light scattering leaves no time-averaged energy in the medium (i.e., the nonlinearity is passive and the excitation is virtual). However, the stimulated Raman effect continues to feed energy into the medium (i.e., the nonlinearity is active and the excitation is real) even when anti-Stokes–Stokes coupling is allowed [Y. R. Shen and N. Bloembergen, Phys. Rev. 137, A1787 (1965)]. This is reflected in the fact that $\chi^{(3)}$ is predominantly real for light-by-light scattering, whereas it is predominantly imaginary for stimulated Raman scattering. This difference manifests itself experimentally in that a dip in Stokes generation occurs at the phased-matched angle in the stimulated Raman effect [R. Y. Chiao and B. P. Stoicheff, Phys. Rev. Letters 12, 290 (1964); E. Garmire, Physics of Quantum Electronics, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Company, Inc., New York, 1966), p. 167; P. V. Avizonis, A. H. Guenther, T. A. Wiggins, R. V. Wick, and D. H. Rank, Appl. Phys. Letters 9, 309 (1966)]. In the present experiment a peak in the generation of the Stokes wave is observed.

[3]P. D. Maker and R. W. Terhune, Phys. Rev. 137, A801 (1965). Recently nonlinear four-photon mixing has been observed in III-V semiconductors [C. K. N. Patel, R. E. Slusher, and P. A. Fleury, Phys. Rev. Letters 17, 1011 (1966)]. Assuming no dispersion, collinear mixing can only give rise to linear gain in the field, as observed by these authors. However, by making the beams noncollinear along the lines indicated in the present paper, it may be possible to obtain exponential gain.

[4]R. Y. Chiao, P. L. Kelley, and E. Garmire, Phys. Rev. Letters 17, 1158 (1966); V. I. Bespalov and V. I. Talanov, Zh. Eksperim. i Teor. Fiz.–Pis'ma Redakt. 3, 471 (1966) [translation: JETP Letters 3, 307 (1966)].

[5]G. Mayer and F. Gires, Compt. Rend. 258, 2039 (1964).

[6]In general, for $\theta < \sqrt{2}\theta_{\rm opt}$, we have

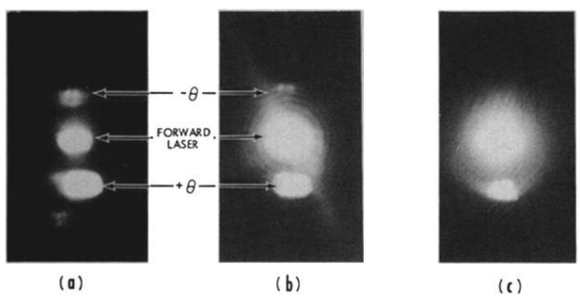
$$I_{-\theta}(L) = I_{+\theta}(L) - I_{+\theta}(0) = I_{+\theta}(0)\frac{k_0^2\theta_{\rm opt}^4}{[g(\theta)]^2}[\sinh\tfrac{1}{2}g(\theta)L]^2,$$

where $g(\theta) = k_0\theta(2\theta_{\rm opt}^2 - \theta^2)^{1/2}$. When $\theta = 0$, this equation becomes

$$I_{-\theta}(L) = I_{+\theta}(0)\frac{k_0^2\theta_{\rm opt}^4L^2}{4},$$

which is the result alluded to in our discussion above,[3] namely, that for collinear beams, the complex amplitudes grow linearly with distance.

[7]P. Lallemand and N. Bloembergen, Phys. Rev. Letters 15, 1010 (1965; C. C. Wang, Phys. Rev. Letters 16, 344 (1966).



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