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BEAM DETERIORATION AND STIMULATED RAMAN EFFECT*

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The most important fundamental discrepancy between theory and experiments in the stimulated Raman effect is that the observed Raman gain is one to two orders of magnitude larger than the theoretical value.[1] The latter is given by[2]

$$g=(2\pi\omega_S^{\,2}/k_{Sz}c^2)|\chi_S''||E_l|^2,$$

where ω_S is the Stokes frequency, k_S the Stokes wave vector, E_l the laser field amplitude, and χ_S'' the resonant Raman susceptibility whose magnitude can be obtained from the spontaneous Raman-scattering data. It was suggested that the observed anomalous gain might be the result of the multimode structure (or hot filaments) of the laser (pumping) beam,[3] but McClung, Wagner, and Weiner, using a nearly single-mode laser beam in the experiments, still found the presence of such an anomalous gain.[1] This, however, does not eliminate the possibility of deterioration of the laser beam into multimodes as the beam interacts with the medium. In this paper, experimental evidence is presented to suggest that scattering mechanisms in a medium can produce inhomogeneities or filamentary structure in an initially homogeneous beam. We believe that these hot filaments are responsible for the many anomalous effects previously observed.

A laser beam, Q switched by cryptocyanine solution and limited in cross section by an aperture in the cavity, was used to generate Stokes radiation in a 20-cm toluene cell (cell A). The laser intensity was varied by a Polaroid prism outside the laser cavity. Another cell (cell B) of variable length, filled with water, benzene, acetophenone, or nitrobenzene, was inserted between the laser and the toluene cell. The threshold of the stimulated Raman scattering was then measured as a function of the length of cell B. The results are shown in Fig. 1. The curves clearly show that the medium in cell B can distort the laser beam in such a way as to help significantly the Raman generation in toluene. Here, the Raman threshold of toluene first increases and then decreases sharply as the length of cell B is increased.

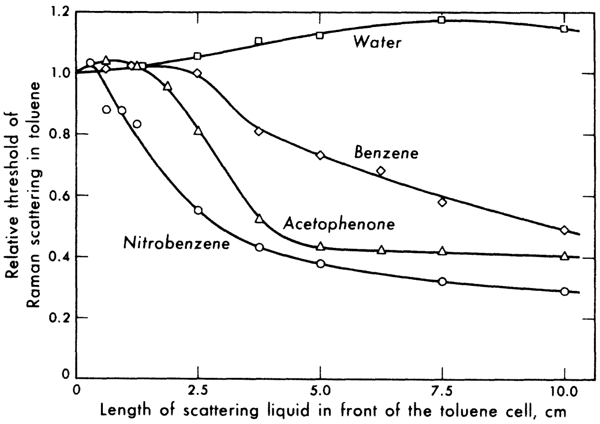

FIG. 1. Raman threshold in toluene versus the cell length of a scattering cell in front of the toluene cell. The scattering cell was filled with water, benzene, acetophenone, or nitrobenzene.

This suggests that the distortion of the beam is due to some kind of stimulated scattering in cell B. The initial rise of the threshold is believed to be the result of insertion loss in cell B. No Raman radiation was generated in cell B, except for the case of nitrobenzene or acetophenone with the cell longer than 3.5 cm. It is therefore reasonable to assert that the beam distortion is due to forward scattering through generation of acoustic and thermal strain (Brillouin and Rayleigh scattering). The maxima and the slopes of these curves show that if forward scattering is responsible for the beam distortion, nitrobenzene should have the largest scattering cross section, followed by acetophenone, benzene, and water. The existing data on incoherent light scattering give the following scattering intensity ratio[4]:

$$I_{\mathrm{NB}}:I_{\mathrm{AC}}:I_{\mathrm{B}}:I_{\mathrm{water}}=10.88:5.66:3.15:0.17.$$

Above threshold, the intensity of the toluene Stokes emission was found to increase appreciably when a 7.5-cm nitrobenzene cell is inserted between the laser and the toluene cell, even though the laser power is somewhat depleted by the generation of Raman emission in nitrobenzene. It was also noticed that the laser beam coming out from a Raman cell was generally less homogeneous than the original beam. Hot, thin laser filaments formed in Raman-active media have been observed by other workers.[5]

That the forward scattering may be responsible for the anomalous Raman gain is also reflected in the temperature effect of the Raman emission. Fig. 2 shows the Raman threshold in nitrobenzene and toluene in a 15-cm cell as a function of temperature. The observed effect is too large to be attributed to the change in the Raman scattering itself. This is confirmed by the fact that when a 7.5-cm nitrobenzene cell was inserted in front of the toluene, the toluene Raman threshold remained more or less constant with temperature. If the temperature of the nitrobenzene cell was varied instead, appreciable change in the toluene Raman threshold was again observed. The curves indicate less beam distortion for higher temperature. This suggests that the forward stimulated Brillouin effect may be the dominating mechanism for beam distortion, since the effect would then be stronger for smaller acoustic damping, and hence for lower temperature.

Theoretically, a laser beam can be distorted or deteriorated into multimodes through nonlinear interaction between light waves and pressure (acoustic) and thermal waves in the medium. The interaction is governed by the set of coupled electromagnetic and acoustic wave equations and the heat diffusion equation. In the limiting case where only the static pressure and thermal strain (electrostriction) are considered, this would lead to the beam-trapping phenomenon proposed by Chiao, Garmire, and Townes.[6] More generally, the initial laser intensity distribution in the frequency and wave-vector space would be broadened a great deal by this mechanism. In other words, a single-mode laser beam can be spoiled into many coherent spatial and temporal modes, which then give rise to hot filaments in the beam and intense spikes in the laser pulse. Usually, the thermal effect is negligible compared to the pressure effect. The forward Brillouin scattering (which includes electrostriction) is possibly responsible for the distortion of the beam. It would have a threshold much lower than the Raman threshold in many media, as seen from the estimate for the case of beam trapping.[6]

Most of the anomalous Raman effects can be explained by the multimode theory.[3] In particular, N coherent laser modes of comparable intensities would give a maximum Stokes gain which is about N times larger than the average gain.[7] The details of the theory will be reported elsewhere.

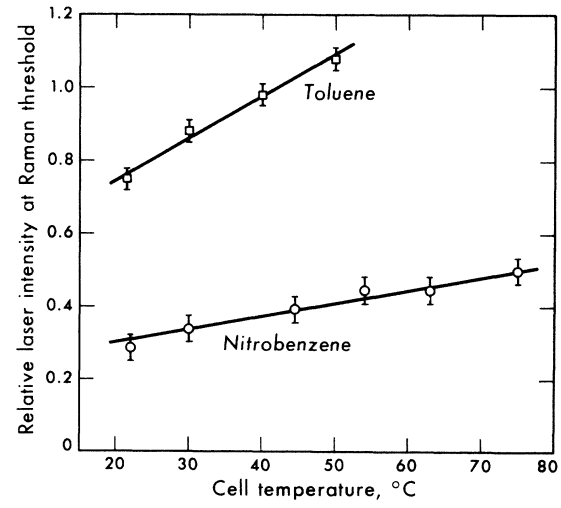

FIG. 2. Raman threshold of toluene and nitrobenzene as a function of temperature.

We would like to thank Professor N. Bloembergen, Dr. F. Quelle, and Professor S. P. Davis for stimulating discussion.

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SELF-FOCUSING OF LASER BEAMS AND STIMULATED RAMAN GAIN IN LIQUIDS*

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Anomalies in the gain of stimulated Raman processes in liquids have been investigated by several groups.[1-3] The study with a Stokes amplifier cell[4] revealed a regime where the gain per unit length was one to two orders of magnitude larger than the value calculated from the spontaneous Raman-emission cross section. The multimode theory of Bloembergen and Shen[5] is inadequate to explain this discrepancy, and the experiment of McClung[6] has shown that the same anomaly exists when the input laser power is essentially in a single mode. The amplifier cell studies[4] suggested that partially depolarized filaments of high intensity are formed as the laser beam passes through the liquid. This self-focusing action of laser beams was foreseen by Chiao, Garmire, and Townes,[7] Askarjan,[8] and Talanof.[8] It is due to the intensity-dependent index of refraction coupled with transverse gradients in the initial intensity distribution. This focusing action should be strongest in liquids with a large quadratic Kerr effect due to anisotropic polarizabilities of the molecules.[9] The purpose of this Letter is to describe new experimental results which support this view and extend the considerations of our very brief previous communication.[10] In the meantime Hauchecorne and Mayer[11] have independently arrived at similar conclusions on the basis of an elegant experiment. Shen and Shaham[12] have also carried out experiments with results similar to ours.

In a conventional Raman oscillator experiment with a collimated laser beam, one can define a threshold condition depending on pump

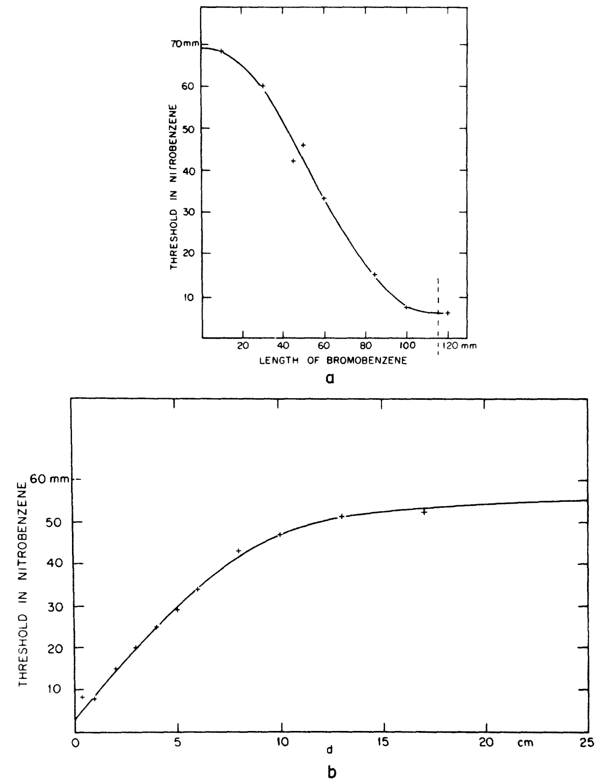

FIG. 1. (a) The threshold length for stimulated Stokes production in a nitrobenzene cell as a function of the length of a cell filled with bromobenzene placed immediately in front. The vertical dashed line indicates the threshold for Stokes production in bromobenzene.
(b) The threshold length for stimulated Stokes production in a nitrobenzene cell preceded by a 30-cm long bromobenzene cell, as a function of the distance between the two cells.