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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

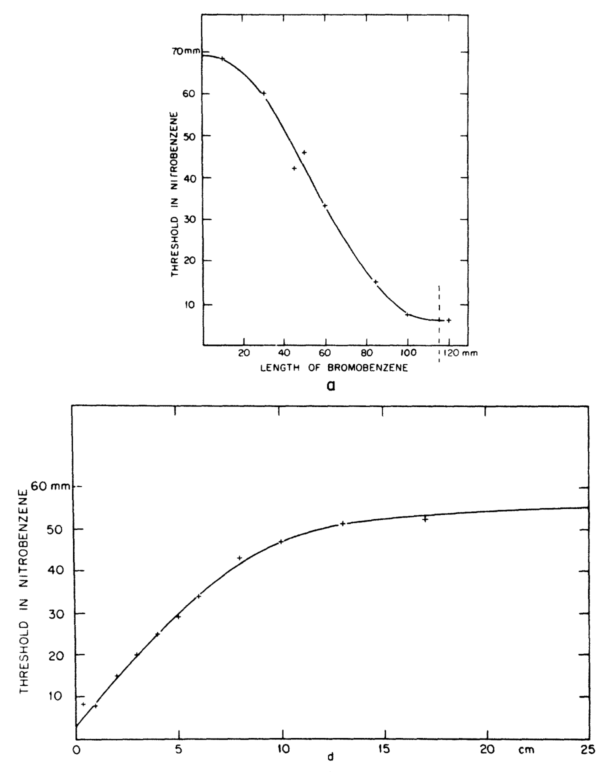

FIG. 1. (a) The threshold length for stimulated Stokes production in a nitrobenzene cell as a function of the length of a cell filled with bromobenzene placed immediately in front. The vertical dashed line indicates the threshold for Stokes production in bromobenzene. (b) The threshold length for stimulated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

intensity and cell length, such that $\mathcal{I}_{th} l_{th}^x$ = constant, where the exponent has the experimental range of value $1 < x < 2$. The threshold constant can be lowered significantly, if the laser beam first traverses a cell with another fluid. Figure 1(a) shows the reduction in the threshold length of a nitrobenzene cell as the length of a cell filled with bromobenzene and placed immediately in front of the nitrobenzene cell is increased. Note that the reduction occurs even before the first cell reaches its Stokes limit. The Stokes shift of the substance in the first cell is far from the nitrobenzene Stokes shift of 1345 cm^{-1}. In addition, filters are used to insure complete decoupling between the cells. The Stokes emission in the first cell may also be suppressed by selective absorbers without affecting the creation of hot filaments. The reduction of a factor 10 in threshold length means that the effective pump intensity is enhanced by almost two orders of magnitude. If the distance between the two cells is increased, the reduction is decreased. The effect of the first cell does not extend beyond 20 cm from its end face, as shown in Fig. 1(b). We have also shown the enhancement of intensity in filaments by means of another nonlinear process, the harmonic production in a quartz platelet placed at the end of the cell. This technique has been used in a more elegant and quantitative manner by Hauchecorne and Mayer.[11]

The creation of the filaments in the first cell has been recorded on a Polaroid film. The difficulty in making them visible for the cell lengths used in our experiments is that the main fraction of the laser intensity goes through without taking part in the focusing process. Since we had previous evidence that the effective pump light is partially depolarized, we put the cell between a crossed polarizer and analyzer to suppress most of the laser light. The image of the end face of the cell is recorded in Fig. 2. When the cell length is not sufficient for self-focusing, a weak homogeneous intensity distribution shown in Figs. 2(a) and 2(b) appears. When self-focusing occurs within the cell length, bright spots appear. Their number increases with increasing cell length. Their diameter is between 20 and 80 μ. This dimension is in agreement with the loss of the filamentary structure due to diffraction in 20 cm of air. The pattern changes from laser shot to laser shot, unless strong reproducible inhomogeneities are introduced into the beam by apertures. Figure 2(e) shows that filaments preferentially form in a circular pattern after diffraction of the beam through a circular hole before entering the cell.

There is a very good correlation between the self-focusing ability of a liquid and its intensity-dependent index of refraction.[9] Liquids are decreasingly effective in the order CS_2, nitrobenzene, bromobenzene, benzene, and acetone, while no detectable focusing action

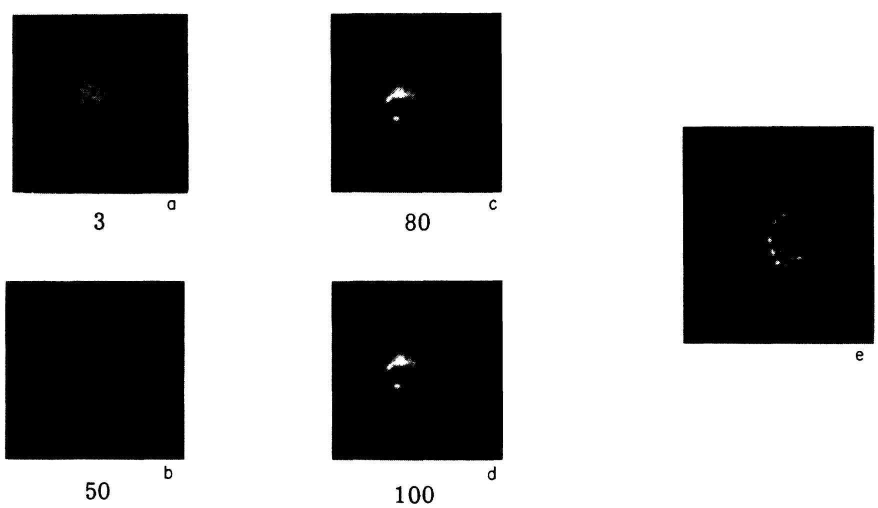

FIG. 2. Photographs of the end face of a nitrobenzene cell, between crossed polarizer and analyzer, of variable length indicated in mm. (a) and (b) The length is below threshold. (c) The length is at threshold for Stokes production. (d) The length is well above threshold. (e) The laser beam is diffracted by a small circular aperture before entering the cell. Filaments are found on rings of maximum transverse intensity gradient.

occurred with isopentane, alcohol, water, and CCl_4. We have found no good correlation with the electrostrictive properties nor with a resonance associated with two-quanta absorption. Theory also predicts that their contribution to the intensity-dependent index should be smaller than that of the anisotropic polarizability.

The experimental threshold for Raman laser action appears to be determined not so much by the value of the Raman susceptibility, but rather by the self-focusing capability of the fluid. This suggests that stimulated Raman oscillations may be induced in liquids that remain below threshold in the unfocused laser beam by mixing them with a focusing liquid. A cell with pure cyclohexane was below threshold in the collimated laser beam. Stimulated Stokes lines of cyclohexane were produced if the cell was filled with a mixture of two-thirds cyclohexane and one-third nitrobenzene. The totally symmetric CCl_4 molecule does not have sufficient focusing action of its own. We have observed three orders of stimulated Stokes light with the characteristic shift of 459 cm^{-1} of the totally symmetric vibration of CCl_4 in a mixture of 75 cc CCl_4, 15 cc nitrobenzene, and 10 cc CS_2. It was necessary to use a mixture of the last two focusing fluids in order to stay below the Stokes threshold of both of these.

The characteristic focusing length follows from the curvature of light rays in a transverse gradient of index of refraction. This length should be smaller than the Fresnel length of the beam, d^2/λ, and than the cell length. If the intensity drops by an amount $\mathcal{I}_0$ over a distance a on either side of an intensity maximum in the beam, the self-focusing length is

$$l_{\text{s.f.}} = \frac{a}{2}\left(\frac{n_0}{2n_2\mathcal{I}_0}\right)^{1/2},$$

where $n_2\mathcal{I}_0$ is the intensity-dependent change in index of refraction. A more detailed calculation of the focusing action in the laser beam has recently been given by Kelley.[13] If one takes a equal to the size of the laser beam d and puts $\mathcal{I}_0$ equal to the intensity at the beam center, he finds that the focusing length under the conditions of our experiments is 1 m for CS_2 with $n_2 = 10^{-11}$ esu; and 3 m for benzene with $n_2 = 0.2\times10^{-11}$ esu. The experimental focusing lengths are an order of magnitude smaller. Furthermore, the whole beam does not focus as a unit. The multimode effect must still be invoked to explain this discrepancy. Apparently random small-scale intensity variations present in the input laser signal are responsible for more rapid and piecemeal focusing of the beam. It is not clear at this moment what factors determine the diameter of the filaments. Apparently some opposing defocusing effects set in after the filaments have reached a given size or intensity.

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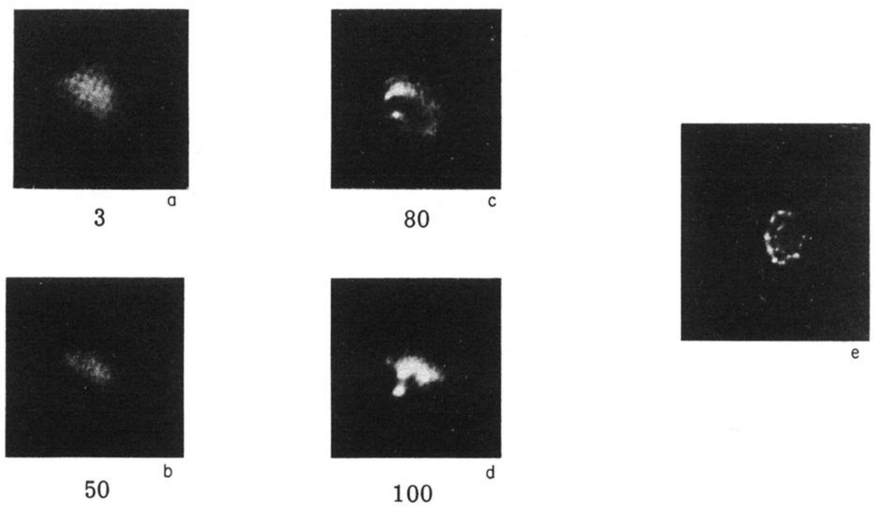

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