of sound measurements very close to the λ line, both at vapor pressure and at higher pressures, is apparent.

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LENGTH-DEPENDENT THRESHOLD FOR STIMULATED RAMAN EFFECT AND SELF-FOCUSING OF LASER BEAMS IN LIQUIDS

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Recent experiments[1-3] have revealed the formation of high-intensity filaments in the laser beam during its passage through Raman-active liquids. Such high-intensity filaments are believed to be formed by the self-focusing action of laser beams,[3,4] which in turn is due to the intensity-dependent index of refraction.[5,6] It has been noted[3] that the experimental threshold for stimulated Raman emission appears to be determined not so much by the value of the Raman susceptibility, but rather by the self-focusing capability of the liquids. In particular, the onset of stimulated Raman emission has been observed to occur only after the beam has traveled some distance through the liquid,[1,3] and it has been proposed[4] that this is the distance required for self-focusing to develop. In this Letter we report our length-dependent threshold data for stimulated Raman emission in several liquids, and show that these data can be interpreted in terms of the predicted dependence of self-focusing. By extrapolating the results to liquid cells of infinite length, we obtain values of critical power for self-trapping[7] of the laser beam in liquids. We believe this is the first measurement from which the critical power for self-trapping has been deduced.

The self-focusing effect can be understood by considering the diffraction of a laser beam in material which exhibits an intensity-dependent index of refraction. For normal dielectrics, the index increases with increasing intensity so that the phase velocity decreases with increasing intensity. A lens effect is thus produced whereby the rays move toward the region of highest intensity and the intensity there increases. This increase in intensity is accompanied by a reduction in effective beam width, and continues until it is limited by other factors. A threshold exists for the onset of self-focusing as it must overcome the spreading of the beam due to diffraction. Chiao, Garmire, and Townes[7] have predicted that a light beam may be trapped at any arbitrary diameter and will thus not spread. They have further predicted that self-trapping occurs at a critical power level independent of the beam diameter. Experiments[3,8] with high-power laser beams have shown that high-intensity filaments of 20 to 80 μ in diameter are formed, presumably due to self-focusing. While it is not yet understood which factors determine the size of the filaments, the distance required for the establishment of these filaments should depend very little on the terminal filament size if the beam is reduced in diameter by an appreciable factor; this is particularly so because the nonlinear nature of the focusing causes the beam diameter to decrease more rapidly the more the beam diameter decreases. This distance has been calculated by Kelley,[4] and has been referred to as the self-focusing length l,

$$l=\frac{n_0}{4}\left(\frac{a^2}{f}\right)\left(\frac{c}{n_2}\right)^{1/2}\Big/[P^{1/2}-P_{\mathrm{cr}}{}^{1/2}]. \tag{1}$$

Here n_0 is the linear index of refraction, c is the speed of light in vacuum, n_2 is related to the dc change in index as defined in Ref. 7, P is the input laser power, and P_{cr} is the critical power for cylindrical beam trapping,[7]

$$P_{\mathrm{cr}}=(1.22\lambda)^2c/256n_2. \tag{2}$$

Equation (1) has been modified slightly from that used by Kelley.[4] Here f is the ratio of the radius of the beam, a, to a characteristic transverse radius of curvature of the laser intensity; it is introduced here as a parameter to allow for deviations from an equiphase Gaussian intensity profile. $f=1$ for such a Gaussian beam assumed by Kelley.[4]

Equation (1) may be rewritten as

$$P^{1/2} = P_{\text{cr}}^{1/2} + \alpha/l, \tag{3}$$

where

$$\alpha = \frac{n_0}{4}\left(\frac{a^2}{f}\right)\left(\frac{c}{n_2}\right)^{1/2}. \tag{4}$$

Equation (3) gives the threshold power for the formation of filaments in a liquid column of a given length. Thus if P is to be taken as the observed threshold for stimulated Raman emission, the plot of $P^{1/2}$ vs $1/l$ for various materials should yield straight lines. The intercepts on the ordinate of these straight lines should give the values for P_{cr}, whereas the slopes of these lines should be proportional to (a^2/f) and could thus be used to determine the ratio f of radii introduced in Eq. (1).

The experimental setup used to obtain the threshold data for stimulated Raman emission has been described elsewhere.[9] The observed Stokes power as a function of the incident laser power was plotted over 10 orders of magnitude in a log-log plot. The value of the threshold laser power was then determined by observing the sharp break in the curve as transition took place from the linear spontaneous region to the stimulated region. The relative accuracy of the threshold values thus obtained is typically ±20% for most liquid cells, and slightly higher for very short cells. The absolute power measurement is estimated to be accurate to ±50%. Figure 1 shows these threshold data for benzene, toluene, and nitrobenzene. One observes that Eq. (1) is well satisfied for all three liquids investigated.

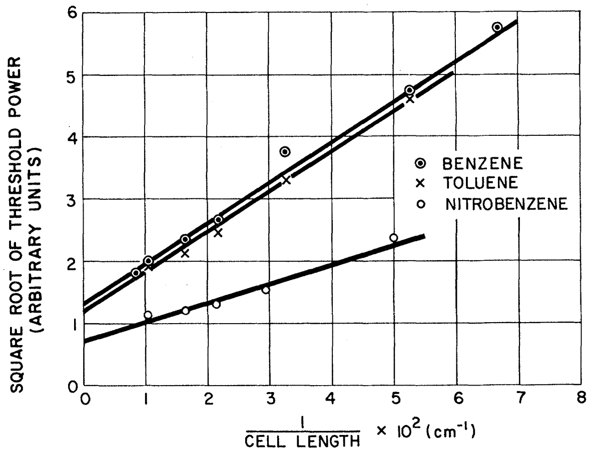

FIG. 1. Plot of the square root of the threshold laser power for stimulated Raman emission as a function of the inverse of the cell length for benzene, toluene, and nitrobenzene.

With the data compiled in Ref. 5 for n_2, the values of P_{cr} calculated from Eq. (2) and from Eq. (2) of Ref. 7 are listed in Table I along with the observed values. Note that the power calculated from our Eq. (2) is one-fourth as much as the critical power deduced in Ref. 7 on the basis of elementary considerations. The observed values fall between these two values.

Also included in Table I are the values of (f/a^2) obtained both in benzene and in nitrobenzene. These two values for (f/a^2) are found to be the same within experimental accuracy. This is to be expected since (f/a^2) is characteristic of the laser beam.

We have studied experimentally the intensity profile of the incident laser beam. The laser beam was found to contain four nearly circular spots approximately equal in intensity and each about 0.1 ± 0.02 cm in diameter. Experiments with two of the spots blocked gave the same results within experimental error. The intensity distribution of these spots was asymme-

Table I. Comparison of critical powers for self-focusing and of ratios f of radii.

Material	Critical power P_{cr} (MW) Observed	Calculated[a]	Calculated[b]	(f/a^2) (cm^2)
Benzene	0.064	0.021	0.085	7.9×10^2
Nitrobenzene	0.019	0.005	0.021	8.3×10^2
Toluene	0.055	...	...	...

[a] Reference 4.
[b] Reference 7.

trical and exhibited curvature smaller than the curvature for a Gaussian beam. Assuming a circular spot, one obtains from Table I values of f near 2. Previously,[3] f has been estimated to be ≈10. Beams whose curvature is greater than the curvature for a Gaussian beam will have larger values of f and thus have shorter self-focusing length. Multimode effects[10] should also increase the f values. On the other hand, possible two-photon absorption tends to impede the self-focusing action, and will reduce the f values.

We have also performed threshold measurements with laser beams of three different cross sections. The result for benzene is plotted in Fig. 2. Here the experimental points are fitted to three straight lines with common intercept on the vertical axis; the slopes of these lines are seen to increase in linear proportion to the cross sectional area of the beam. This is in agreement with the predictions that the critical power for self-trapping is independent of the beam cross section, and that the quantity $(P^{1/2}-P_{\mathrm{cr}}^{1/2})$ expressing the threshold for Raman laser action should increase as the square of the radius of the beam. Qualitative results of a similar nature have also been observed by McClung.[11]

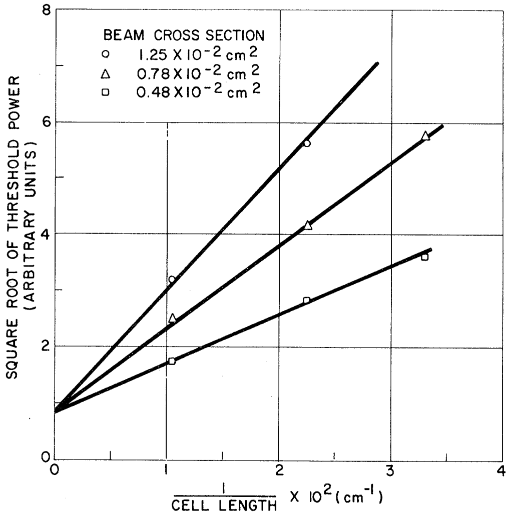

FIG. 2. Same plot as Fig. 1 for benzene, but with beams of different cross sections. The beam cross section used is (a) 1.25×10^{-2} cm^2, (b) 0.78×10^{-2} cm^2, and (c) 0.48×10^{-2} cm^2. This corresponds to a ratio of 1.6:1:0.615 among the cross sections, as may be compared with the ratio of 1.5:1:0.6 among the observed slopes.

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