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Raman-limited beam diameters in the self-focusing of laser light

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Numerical calculations show that high conversion of laser light into stimulated Raman light in the *forward* direction limits the beam diameter of the self-focused light. The dependence of the limiting diameter on material parameters and experimental conditions is calculated. In the experiments we used single-frequency laser pulses of $\sim 10^{-8}$-sec duration. Our measured minimum diameters in liquid benzene, mixtures of benzene and CS_2, and toluene are in fair agreement with values calculated from our steady-state theory. The rapid movement of the focus at the end of the liquid cell gives rise to transient stimulated Raman scattering or transient self-focusing in liquids with long relaxation times. In CS_2, nitrobenzene, and bromobenzene these transient effects have to be taken into account in the comparison between calculated and measured limiting diameters.

I. INTRODUCTION

Self-focusing of laser pulses in liquids has received considerable attention.[1,2] Many aspects of this phenomenon appear to be well understood. However, one of the interesting problems, the origin and size of the limiting diameters, is still under discussion. Several mechanisms have been suggested for the explanation of the limiting diameters. Saturation of the orientational Kerr effect[3,4] has been found to be inadequate to explain the experimental results. Therefore, steric effects have been included in the calculations of the Kerr constant.[5] Multiphoton absorption of the laser light has been discussed as a possible limiting mechanism.[6,7] It is difficult to estimate the beam diameter limited by multiphoton absorption, since numbers for the nonlinear absorption coefficients are not known for most liquids. Avalanche ionization was also suggested to stabilize the size of the self-focused light beam.[8] The estimated limiting diameters are in the range of the experimentally observed values in liquids. More recently, the depletion of the laser pulse near the focal region by *backward* stimulated Raman scattering has been proposed to cause a limitation of the diameter of the self-focused light.[9] The correct order of magnitude of the limiting diameters in liquids was obtained from these calculations.

Although these models suggest the existence of limiting diameters, calculations considering the details of the limiting mechanisms and the self-focusing action are still lacking.

In this paper experimental and theoretical evidence is presented by the authors that *forward* stimulated Raman scattering (SRS) limits the beam diameter of the self-focused light in several liquids. In contrast to the models above, computer calculations were carried out taking into account the details of the self-focusing action and of stimulated Raman scattering for the steady-state case. Quantitative comparison with our experimental results is made.

In Sec. II the steady-state theory of Raman-limited minimum diameters is outlined and numerical results of computer calculations are given. The effect of the moving focus on the limiting diameters in liquids with long vibrational and reorientational relaxation times is discussed qualitatively. In Sec. III the experimental results on several pure and mixed liquids are described. Comparison is made with the theoretical predictions. A short summary of the results of this paper is given in Sec. IV.

II. THEORY

The usual treatment of self-focusing of light starts with the wave equation for the light field $\vec{E}$,[1]

$$\nabla^2\vec{E} - \frac{\epsilon_0}{c^2}\frac{\partial^2\vec{E}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2\vec{P}^{NL}}{\partial t^2}. \tag{1}$$

Here c is the velocity of light in vacuum and ϵ_0 the intensity-independent part of the dielectric constant. The nonlinear polarization is given by

$$\vec{P}^{NL} = \frac{\Delta\epsilon}{4\pi}\vec{E} + N\frac{\partial\alpha}{\partial\overline{q}}\,\overline{q}\vec{E}. \tag{2}$$

The first term on the right-hand side contains the intensity-dependent part $\Delta\epsilon$ of the dielectric constant, which leads to the self-focusing action. Usually, only this term is included in the calculations. Since we are interested in the effect of stimulated Raman scattering on the self-focusing of light,[10] we added the second term, which describes the change of polarizability α with vibrational coordinate $\bar{q}$. N is the number of molecules per cm^3.

The molecular vibrations (damping constant Γ_R and resonance frequency ω_0) are driven by the light field,

$$\frac{\partial^2\bar{q}}{\partial t^2}+\Gamma_R\frac{\partial\bar{q}}{\partial t}+\omega_0^2\bar{q}=\frac{1}{2m}\frac{\partial\alpha}{\partial\bar{q}}\vec{\mathrm{E}}^2 . \qquad (3)$$

Here m is the mass associated with the $\bar{q}$ vibration.

In the liquids investigated in this paper the optical Kerr effect gives the major contribution to the nonlinear dielectric constant. $\Delta\epsilon$ is described by a relaxation equation of the following form:

$$\frac{\partial(\Delta\epsilon)}{\partial t}+\frac{1}{\tau_K}\Delta\epsilon=\frac{\epsilon_2}{\tau_K}\vec{\mathrm{E}}^2 . \qquad (4)$$

For light pulses long compared to the relaxation time τ_K of the orientational Kerr effect, the steady-state value

$$\Delta\epsilon=\epsilon_2\vec{\mathrm{E}}^2 \qquad (5)$$

is obtained from Eq. (4). ϵ_2 is the nonlinear part of the dielectric constant. Using the relation $\epsilon=n^2$ between the dielectric constant and the refractive index, we get $\epsilon_2=2n_0n_2$. n_0 and n_2 are the linear and nonlinear part of the refractive index, respectively.

For the solution of the coupled equations (1)–(4) the light fields are assumed to be linearly polarized waves with slowly varying amplitudes. We consider only the laser field E_L and the first Stokes line E_S. k_L, k_S, ω_L, and ω_S are the magnitudes of the wave vectors and frequencies of the laser and Stokes light. This leads to the following equations for the light fields:

$$\frac{\partial^2E_L}{\partial r^2}+\frac{1}{r}\frac{\partial E_L}{\partial r}+2ik_L\left(\frac{\partial E_L}{\partial z}+\frac{\epsilon_0^{1/2}}{c}\frac{\partial E_L}{\partial t}\right)=-k_L^2\frac{\Delta\epsilon}{\epsilon_0}E_L-2k_L\frac{\pi N\omega_L}{\epsilon_0^{1/2}c}\frac{\partial\alpha}{\partial q}E_Sq , \qquad (6)$$

$$\frac{\partial^2E_S}{\partial r^2}+\frac{1}{r}\frac{\partial E_S}{\partial r}+2ik_S\left(\frac{\partial E_S}{\partial z}+\frac{\epsilon_0^{1/2}}{c}\frac{\partial E_S}{\partial t}\right)=-k_S^2\frac{\Delta\epsilon}{\epsilon_0}E_S-2k_S\frac{\pi N\omega_S}{\epsilon_0^{1/2}c}\frac{\partial\alpha}{\partial q}E_Lq^* , \qquad (7)$$

for the amplitude of the molecular vibrations,

$$\frac{\partial q}{\partial t}+\tfrac{1}{2}\Gamma_R q=\frac{i}{4m\omega_0}\frac{\partial\alpha}{\partial q}E_LE_S^* , \qquad (8)$$

and for $\Delta\epsilon$,

$$\frac{\partial(\Delta\epsilon)}{\partial t}+\frac{\Delta\epsilon}{\tau_K}=\frac{\epsilon_2}{2\tau_K}(|E_L|^2+|E_S|^2) . \qquad (9)$$

It has been taken into account that $\Delta\epsilon$ cannot follow terms varying on a time scale very short compared to τ_K.

First, Eqs. (6)–(9) will be solved for the steady-state case; then, the influence of transient SRS and transient Kerr effect on the limiting diameters of the self-focused light will be discussed.

A. Steady-state self-focusing and Raman scattering

1. *Differential equations*

When the molecular vibrations are heavily damped and the relaxation time τ_K is very short, i.e.,

$$\tfrac{1}{2}\Gamma_R q\gg\frac{\partial q}{\partial t}\ \text{and}\ \frac{\Delta\epsilon}{\tau_K}\gg\frac{\partial(\Delta\epsilon)}{\partial t} ,$$

Eqs. (8) and (9) are readily solved for q and $\Delta\epsilon$. Inserting the results into (6) and (7), we obtain the coupled steady-state equations for E_L and E_S[11]:

$$\frac{\partial^2E_L}{\partial r^2}+\frac{1}{r}\frac{\partial E_L}{\partial r}+2ik_L\frac{\partial E_L}{\partial z}=-\frac{\epsilon_2}{2\epsilon_0}k_L^2(|E_L|^2+|E_S|^2)E_L-ig_L\frac{c\epsilon_0^{1/2}k_L}{8\pi}|E_S|^2E_L , \qquad (10)$$

$$\frac{\partial^2E_S}{\partial r^2}+\frac{1}{r}\frac{\partial E_S}{\partial r}+2ik_S\frac{\partial E_S}{\partial z}=-\frac{\epsilon_2}{2\epsilon_0}k_S^2(|E_L|^2+|E_S|^2)E_S+ig_S\frac{c\epsilon_0^{1/2}k_S}{8\pi}|E_L|^2E_S . \qquad (11)$$

The important terms are on the right-hand side of Eqs. (10) and (11). The first terms lead to the self-focusing of the Stokes and laser light. Note that both the laser and the Stokes intensity contribute to the increase of the dielectric constant. The second term in Eq. (10) describes the attenuation of the laser light by SRS with a loss factor

$$g_L=\frac{8\pi^2\omega_LN}{\epsilon_0c^2m\Gamma_R\omega_0}\left(\frac{\partial\alpha}{\partial q}\right)^2=\frac{8\pi cN\omega_L}{\hbar\omega_S^4\epsilon_0\delta\bar{\nu}_R}\frac{d\sigma}{d\Omega} . \qquad (12)$$

Here the linewidth $\delta\bar{\nu}_R$ and the scattering cross section $d\sigma/d\Omega$ of spontaneous Raman scattering have been introduced. The second term in Eq. (11) corresponds to the generation of Raman light with a gain factor

$$g_S = \omega_S g_L/\omega_L\,. \tag{13}$$

The loss of the laser light is larger than the gain of the Stokes light. The difference in energy is used for the excitation of molecular vibrations.

For the numerical solution of Eqs. (10) and (11) the following normalized quantities are introduced: radial distance $\bar{r}=r/a_L$, axial distance $\bar{z}=z/2k_La_L^2$, electric field $\bar{E}_{L,S}=(\epsilon_2/2\epsilon_0)^{1/2}k_La_LE_{L,S}$, gain factor $\bar{g}=g_Sc\epsilon_0^{3/2}/4\pi k_S\epsilon_2$, and $\bar{\kappa}=k_S/k_L$. Here a_L is the $1/e$ radius of the Gaussian intensity distribution of the incoming laser beam and $k_La_L{}^2$ is the diffraction length of the light beam. With these dimensionless quantities, Eqs. (10) and (11) have the following form:

$$i\frac{\partial\bar{E}_L}{\partial\bar{z}}+\frac{\partial^2\bar{E}_L}{\partial\bar{r}^2}+\frac{1}{\bar{r}}\frac{\partial\bar{E}_L}{\partial\bar{r}}+(|\bar{E}_L|^2+|\bar{E}_S|^2)\bar{E}_L+i\bar{g}|\bar{E}_S|^2\bar{E}_L=0, \tag{14}$$

$$i\bar{\kappa}\frac{\partial\bar{E}_S}{\partial\bar{z}}+\frac{\partial^2\bar{E}_S}{\partial\bar{r}^2}+\frac{1}{\bar{r}}\frac{\partial\bar{E}_S}{\partial\bar{r}}+\bar{\kappa}^2(|\bar{E}_L|^2+|\bar{E}_S|^2)\bar{E}_S-i\bar{g}\bar{\kappa}^2|\bar{E}_L|^2\bar{E}_S=0. \tag{15}$$

2. *Calculation of limiting diameter*

Equations (14) and (15) are transformed into difference equations and solved numerically on a computer for practical values of the Raman gain factor ($\bar{g}$) and the Raman frequency shift ($\bar{\kappa}$). In the calculations the following initial conditions are used: Gaussian intensity distribution of the laser and Stokes light; the diameter and divergence of both beams were assumed to be equal; the ratio of Raman and laser intensity was estimated from spontaneous scattering data. The beam divergence D_V at the entrance of the medium ($z=0$) was taken into account in the calculations in the same way as in Ref. 12; we define $D_V=\Delta a/l$, with the normalized value $\bar{D}_V=D_V(\frac{1}{2}k_La_L)$. l is the length of the medium and Δa the difference of the $1/e$ radii of the laser light at distance $z=0$ and $z=l$ in a medium without self-focusing. From the calculations we obtain the intensity distribution, the peak intensity, and the diameter of the laser and Stokes beam as a function of distance z.

In the following figures the normalized quantities defined above are used at the lower scale and the left-hand scale. The numbers at the upper scale, the right-hand scale, and in brackets correspond to a light beam of wavelength $\lambda_L=6943$ Å with a $1/e$ radius $a_L=261$ μm incident on CS_2.

In Fig. 1 typical results of our computer calculations are shown.[13] The normalized diameters of the laser and Stokes beams are plotted as a function of normalized distance $\bar{z}$ near the end of the medium [solid and broken line in Fig. 1(a), respectively]. They decrease continuously with increasing $\bar{z}$ and reach their minimum values approximately at the same distance $\bar{z}\simeq\bar{l}$. The dotted line corresponds to the case when stimulated Raman scattering is neglected in the calculations. The figure shows quite vividly that the diameter of the laser beam continues to decrease without SRS.

In Fig. 1(b) the normalized intensities of the laser and Stokes light in the center of the beam are shown (solid and broken line, respectively). First, the laser intensity increases due to the self-focusing action and the Stokes light is amplified by stimulated Raman scattering. When the conversion efficiency of laser into Raman light exceeds approximately 15%, the laser intensity begins to decrease due to the generation of Stokes light. It should be emphasized that the minimum diameter of the laser light [see Fig. 1(a)] occurs close to the position where the laser intensity has its maximum [Fig. 1(b)]. When we neglect SRS

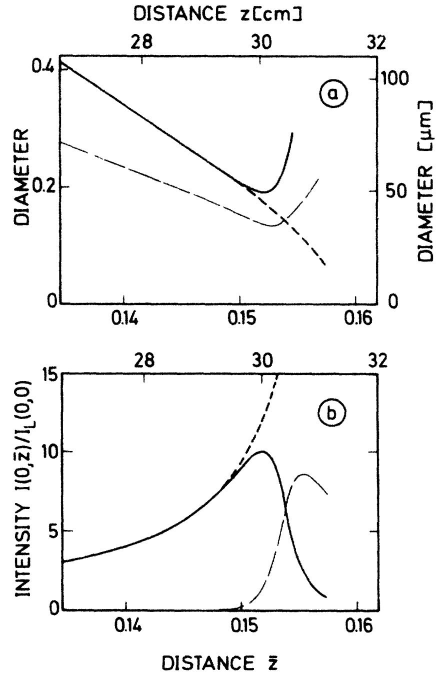

FIG. 1. (a) Normalized beam diameter of laser and Stokes light (solid and broken line, respectively) versus normalized propagation distance $\bar{z}$ in the medium. Dotted line: beam diameter of laser light without stimulated Raman scattering. (b) Normalized on-axis intensities of laser and Stokes light: $\bar{g}=3.0$; $\bar{E}_L(0,0)=5.3$ ($P_L=100$ kW); $\bar{E}_S(0,0)=2.3\times10^{-6}$ ($P_S=2\times10^{-8}$ W); $\bar{\kappa}=0.95$; $\bar{D}_V=1.0$ ($D_V=5\times10^{-4}$ rad in CS_2).

in our calculations the laser intensity continues to rise with distance (dotted line) because of the self-focusing action.[4,14]

For an explanation of the minimum diameters, three different effects have to be considered: (i) the loss of light energy due to the excitation of molecular vibrations; (ii) the strong diffraction of Stokes light out of the center of the light beam; (iii) the depletion of the laser intensity in the center of the beam due to the conversion into Raman light. The minimum diameters of the laser and Stokes light will be discussed separately.

a. Laser light. The main cause for the limiting diameter of the laser light is point (iii). Because of the high conversion efficiency, the depletion of the laser intensity in the center of the beam is larger than its increase due to the self-focusing action. In order to illustrate this point the calculated intensity distributions of the light beams versus normalized radial coordinate $\bar{r}$ are plotted in Fig. 2 (near the minimum diameter, i.e., at $\bar{z} \simeq \bar{l}$).[13] When SRS is taken into account in the calculations (solid line), the peak intensity in the center of the laser beam ($\bar{r}=0$) is smaller than without SRS (dotted line). Since the conversion efficiency depends strongly on the laser intensity, almost no SRS occurs in the wings of the beam. It is seen from Fig. 2 that the depletion of the laser intensity in the center of the beam leads to an increased beam diameter. With rising conversion efficiency the laser intensity is substantially reduced and finally decreases with propagation distance $\bar{z}$ [see maximum in Fig. 1(b)] leading to the minimum value of the beam diameter [see Fig. 1(a)].

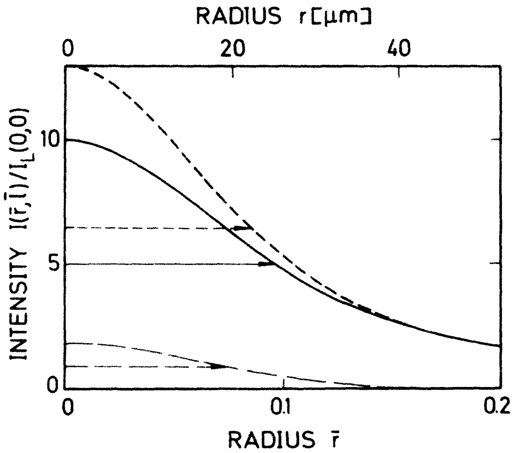

FIG. 2. Laser intensity with and without Raman scattering (solid and dotted line, respectively) and Stokes intensity (broken line) versus radial distance $\bar{r}$ near minimum diameter ($\bar{z} \simeq \bar{l}$). Parameters of the calculations; see Fig. 1.

It should be noted that the final size of the limiting diameter is also influenced by points (i) and (ii). The loss of light intensity by the excitation of molecular vibrations and the diffraction of Stokes light (note the small diameter in Fig. 2) reduce the nonlinear part of the dielectric constant and, therefore, the self-focusing action.

b. Stokes light. The diameter of the Stokes beam decreases with propagation distance $\bar{z}$ for two reasons. First, it is focused by the nonlinear dielectric constant. Second, the gain is highest in the center of the laser beam; therefore, the diameter of the Raman light beam is smaller than that of the laser light and decreases with $\bar{z}$.[15] For high conversion efficiency the nonlinear dielectric constant decreases [see above, points (i) and (ii)] and the Raman gain in the center of the beam begins to saturate [point (iii)] leading to a reduction of the focusing action. The strong diffraction of the Stokes light overcomes the focusing and a minimum diameter is observed for the Stokes light.

Note that we considered only the first Stokes line in the calculations assuming that higher-order Stokes lines are of minor importance. This is justified since the limiting diameters occur at a conversion efficiency of ~15% (laser to first Stokes line) where the second Stokes intensity is small compared to the first Stokes intensity.[16]

3. *Dependence of limiting diameter on material parameters and experimental conditions*

We discuss now the dependence of the limiting diameter on material parameters, e.g., nonlinear dielectric constant ϵ_2, Raman gain factor g_S, and experimental conditions, e.g., incident laser power P_L, length l of the medium, initial intensity $I_S(0,0)$ of the Raman light, divergence D_V of the laser beam.

a. Raman gain. For an explanation of the results it should be recalled that the limiting diameter occurs at high conversion of laser into Raman light. The conversion efficiency depends critically on the Raman gain

$$G_0 = g_S I_L(0,0)l = g_S P_L l / a_L^2 \pi , \tag{16}$$

where $I_L(0,0)$ is the incident laser intensity in the center of the beam. In normalized quantities we get

$$G_0 = 2\bar{\kappa}\bar{g}\bar{E}_L{}^2(0,0)\bar{l} . \tag{17}$$

When the Raman gain G_0 is low, we expect small values of the limiting diameter. For a larger value of G_0 the Stokes light reaches its high conversion efficiency at a shorter distance in the liquid, where the beam diameter is still larger.

Therefore, the limiting diameter increases with Raman gain G_0. In fact, it will be seen below (Fig. 7) from the results of the numerical calculations that the limiting diameter rises strongly with the normalized gain factor $\bar{g}$, which is proportional to G_0 [see Eq. (17)].

b. Initial Stokes intensity. We investigated the influence of the initial conditions of the Stokes light on the limiting diameter.[13] The limiting diameters of the laser and Stokes light, d_L and d_S, respectively, rise slowly when the Stokes intensity $I_S(0,0)$ at the entrance of the medium increases over several orders of magnitude (see Fig. 3). For larger values of the initial Stokes intensity a shorter distance in the medium is necessary to obtain high conversion efficiency. The shorter distance corresponds to a larger beam diameter.

c. Laser-beam divergence. In Fig. 4 the limiting diameters of the laser and Stokes light are plotted versus normalized divergence $\bar{D}_V$ of the laser beam at the entrance of the medium.[13,17] The incident laser power was chosen in such a way that the limiting diameter occurred at the same distance $\bar{l}$ independent of $\bar{D}_V$. For a divergent light beam a higher threshold power is needed than for a parallel beam to obtain self-focusing at the same length.[1] A higher laser power corresponds to a larger Raman gain [see Eq. (16)] and, therefore, to a larger value of the limiting diameter. The limiting diameter increases with divergence $\bar{D}_V$ (see Fig. 4).

d. Length of medium or laser power. In Fig. 5 the calculated laser beam diameter[13] is shown as a function of propagation distance $\bar{z}$ for different values of the incident laser power P_L (curves 1, 2, and 3). With increasing power the minimum diameter occurs at shorter length $\bar{l}$. In the insert of Fig. 5 the limiting diameter $\bar{d}_L$ of the laser light is plotted versus length $\bar{l}$. It decreases rapidly with increasing length $\bar{l}$ and rises slowly after a minimum value. A qualitative explanation is obtained as follows. Since the limiting diameter is small, it occurs at a length l which is approximately equal to the self-focusing length l_f.[18] From previous calculations[4] the relation between the laser power P_L and l_f is known,

$$P_L/P_{cr} = (0.858 + 0.369\, k_L a_L^2/l_f)^2 . \qquad (18)$$

Here P_{cr} is the critical power for self-focusing. For short length l ($\simeq l_f$) we have $P_L \propto 1/l^2$; according to Eq. (16) we get $G_0 \propto 1/l$, i.e., we expect a large Raman gain and a large limiting diameter. If the length of the medium is large, P_L is approximately constant [See Eq. (18)] and the gain $G_0 \propto l$ is again large. For intermediate values of l the Raman gain has a minimum, explaining the minimum of the limiting diameter.

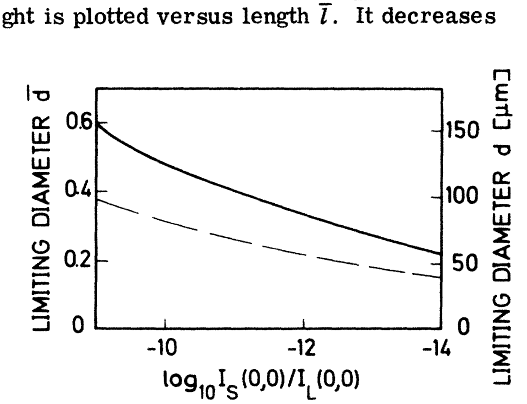

FIG. 3. Limiting diameters $\bar{d}$ of laser and Stokes light (solid and broken line, respectively) versus normalized Stokes intensity $I_S(0,0)/I_L(0,0)$ at the entrance of the medium. Parameters of the calculations; see Fig. 1.

B. Transient self-focusing and Raman scattering

The time duration of our laser pulses (~15 nsec) is long compared to the dephasing time $\tau_R = 1/\Gamma_R$ of the molecular vibrations and the orientational relaxation time τ_K of the Kerr effect (several psec; see Table II). Nevertheless, SRS and self-focusing are transient phenomena in several liquids. The reason is that for a time-dependent incident laser power $P_L(t)$ the focus moves quite rapidly through the medium during the self-focusing process.[19]

We calculate the velocity v_f of the focus by solving Eq. (18) for the self-focusing length l_f and differentiating with respect to time. We get

$$v_f = \left| \frac{dl_f}{dt} \right| = \tfrac{1}{2} l_f \frac{1}{1 - 0.858 (P_{cr}/P_L)^{1/2}} \frac{1}{P_L} \frac{dP_L}{dt} . \qquad (19)$$

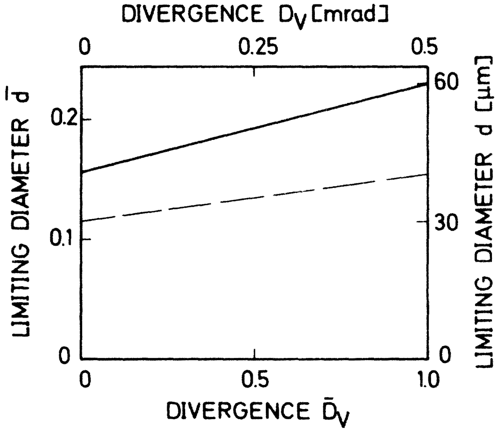

FIG. 4. Limiting diameters $\bar{d}$ of laser and Stokes light (solid and broken line, respectively) versus beam divergence $\bar{D}_V$: $\bar{g} = 3.0$; $\bar{E}_L(0,0) = 5.3$ for $\bar{D}_V = 1$; $\bar{E}_L(0,0) = 3.86$ for $\bar{D}_V = 0$; $\bar{E}_S(0,0) = 2.3 \times 10^{-6}$; $\bar{\kappa} = 0.95$.

Using typical numbers for our experimental situation $l_f = 30$ cm, $P_L/P_{cr} \simeq 5$, and

$$\left(\frac{dP_L}{dt}\right)\left(\frac{1}{P_L}\right) \simeq 0.2 \text{ nsec}^{-1},$$

a value of $v_f \simeq 5\times 10^9$ cm/sec is obtained.

When Δl_f is the length of the focal region,[20] a material element experiences the high intensity of the self-focused light for a time duration of $\Delta t_f = \Delta l_f / v_f$. This time will be called transit time of the focus through a volume element. For the quasi-steady-state situation, Δl_f is obtained from our computer calculations discussed in the preceding sections. We found that the length of the focal region depends strongly on the size of the limiting diameter. $\overline{\Delta l}_f$ increases from 2×10^{-4} to 3.9×10^{-3} ($\Delta l_f = 0.072$ to 1.4 cm) when the limiting diameter varies from $\bar{d}_L = 0.017$ to 0.123 ($d_L = 6$ to 43 μm). Here, Δl_f and d_L are obtained from their normalized quantities using the measured $1/e$ radius $a_L = 350$ μm (see Sec. III C 1). Since Δl_f is small and the focal spot moves with high velocity v_f, the transit time Δt_f is very short, 14 and 280 psec for our examples, respectively. When the relaxation time is of the order of these transit times a transient behavior of SRS and the optical Kerr effect is expected in the focal region.

We discuss now the effect of *transient SRS* on the limiting diameter for quasistationary self-focusing of light. One of the important characteristics of transient stimulated scattering of light is the reduced gain compared to the steady state.[21] For a smaller Raman gain higher light intensity and, therefore, a smaller beam diameter is necessary to obtain high-conversion efficiency of laser into Raman light. As a result a smaller limiting diameter is predicted for transient Raman scattering. It is difficult to calculate quantitatively the reduction of the limiting diameter since in our experimental situation the transient behavior is confined to the end of the medium where the focal spot moves with high velocity. In the major part of the medium the intensity varies on a nanosecond time scale justifying a quasi-steady-state description. Quantitative results of the transient case may be obtained by simultaneous solution of Eqs. (5)–(8), which requires an enormous amount of computer time.

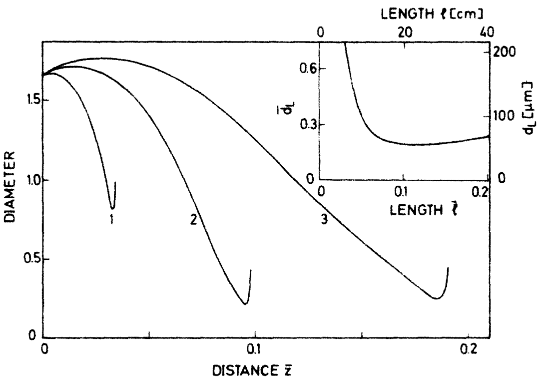

FIG. 5. Normalized beam diameter of the laser light versus distance $\bar{z}$. Curve 1: $\bar{E}_L(0,0) = 11.8$ ($P_L = 496$ kW); curve 2: 6.5 (141 kW); curve 3: 4.8 (82 kW). Insert: Limiting diameter $\bar{d}_L$ of the laser light versus length $\bar{l}$. Parameters of the calculations; see Fig. 1.

The situation is similar for *transient self-focusing*.[7,22] Here Eqs. (6)–(9) have to be solved (on a computer), assuming a steady-state behavior of SRS. Qualitatively it is clear from Eq. (9) that in the transient situation the intensity-dependent increase $\Delta\epsilon$ of the dielectric constant is less than in the steady-state case. This is equivalent to a smaller nonlinearity ϵ_2 and a larger value of $\bar{g}$. We predict, therefore, an increase of the Raman-limited beam diameter when the Kerr effect is transient. Preliminary calculations based on the paraxial-ray theory indicate that the limiting diameters for transient self-focusing are indeed larger than the steady-state values.[23] When both SRS and self-focusing are in the transient regime[7] it is difficult to estimate the limiting diameter without solving the corresponding differential equations.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Experimental setup

In the experiments we used a giant-pulse ruby laser described in Ref. 12. The laser emitted a light beam with a single frequency (width $<10^{-2}$ cm^{-1}) and a Gaussian intensity distribution over the cross section. The peak power was 500 kW and the time duration 15 nsec.

In Fig. 6 the experimental setup for measuring the limiting diameters of the self-focused light is shown. The diameter of the incident laser beam is reduced by a telescope (Li1 and Li2) to approximately $d_0 = 0.6$ mm at the entrance window of the liquid cell C. The length of the cell was $l = 30$ cm. Part of the laser light is coupled out by a glass plate G1 and measured with photocell Ph1. Photocell Ph2 measured the backscattered stimulated Raman power.

Two different methods were used to determine the limiting diameter at the exit window of the liquid cell:

(a) A microscope Mi (resolution 1.5 μm) produced magnified images of the exit window on photographic plates R and L [see Fig. 6(a)]. Filters Fi1 and Fi2 transmit only the Stokes and laser light, respectively. The intensity distributions and beam diameters of the Raman and laser light were obtained from microdensitometer traces of the photographic plates R and L. Special care was taken to calibrate the plates properly;

(b) No microscope was used in the experiments. A photographic plate P was immersed in the liquid (for a short time) and exposed directly by the self-focused light near the end of the cell [see Fig. 6(b)]. The limiting diameter was obtained by analyzing the plate with a microscope under high magnification.

The photographic plates P ("high-resolution plates" from Kodak) were chosen for the following important properties: high spatial resolution (~2000 lines/mm); low sensitivity at the laser frequency, because the light intensity in the focus is high. We found a linear density curve of the plates for short times of development. The sensitivity at the Stokes frequency was measured to be several orders of magnitudes lower than that at the laser frequency.

The advantages of method (b) are evident: high resolution, no problems with adjustment, recording of the total beam cross section. The following drawbacks should be mentioned. A quantitative determination of the beam diameters was not possible in our case, because no microdensitometer was available for measuring the small diameters (several μm). No filters can be used for attenuation or frequency selection without disturbing the self-focusing action. Considering these facts, method (b) was used for a quick survey, while quantitative measurements of the limiting diameters were made with method (a).

B. Limiting diameters

In our investigations the limiting diameter was determined in the following way: We measured the diameter of the laser beam at the end of the cell as a function of laser power (for different laser shots). First, the beam diameter decreases with increasing laser power.[12] If a definite laser power, the threshold power for self-focusing, is exceeded, the diameter remains approximately constant. The corresponding value is called the minimum or limiting diameter. It is important to note that strong stimulated Raman scattering is observed only when the limiting diameter is reached.

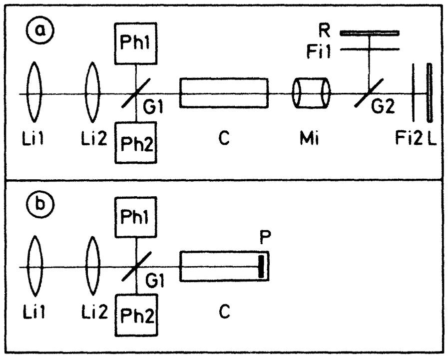

FIG. 6. Schematic of experimental system. For details see text.

The measured minimum diameters for the laser light are listed in Table I for benzene, mixtures of benzene and CS_2, nitrobenzene, bromobenzene, toluene, and CS_2. Mixtures of benzene and CS_2 were investigated because the limiting diameter depends strongly on the concentration of CS_2. It should be noted that the addition of only 7% and 14% CS_2[24] to benzene reduced the limiting diameter of benzene to 24 and 16 μm, respectively.

With the experimental setup shown in Fig. 6(a) we observed the limiting diameters both in the laser and Raman light when appropriate filters Fi1 and Fi2 were used. Usually the diameters of the Raman light were somewhat smaller than the limiting diameters of the laser light. It should be emphasized that the conversion of laser light into forward Raman light was high (>10%). This result was obtained by comparing the densities of the photographic plates [R and L in Fig. 6(a)] and taking into account the filters and different sensitivities of the plates at the Stokes and laser frequency. The measured high conversion efficiency is very important for the comparison with the computer calculations (see below).

C. Discussion

Before a comparison is made between our experimental results and our calculations the following points should be noted. In the calculations the *intensity* distribution of the light waves is obtained and the diameter is taken at the half-maximum points. In the experiments the *energy*

TABLE I. Measured limiting diameters d_L of the laser light. The numbers are average values of several laser shots. The diameters have not been corrected for the resolution of the microscope (~1.5 μm).

	d_L (μm)
Benzene	45
Benzene-CS_2	
93%-7%	24
86%-14%	16
Nitrobenzene	15
Bromobenzene	15
Toluene	10
CS_2	6

density distribution and its diameter is measured, because the photographic plate is a time-integrating device. We have shown[25] that the deviations between the diameters of the intensity and energy density distributions are usually small. In the following discussion the measured diameters will be compared directly with those taken from the computer calculations.

1. *Steady state*

First, the limiting diameters of benzene, mixtures of benzene and CS_2, and toluene will be discussed. The steady-state theory is believed to be a good approximation in these investigations, because the relaxation times τ_R and τ_K are very short (several psec; see Table II). The determining quantity in our calculations is the dimensionless gain factor $\bar{g}$, which is proportional to the Raman gain factor g_S and to $1/n_2$, the nonlinear refractive index. For a comparison of our experiments with the calculations the values of g_S and n_2 must be known. We determined n_2 from measurements of the threshold power P_L^{th} for self-focusing (using the known relation[4] between n_2 and P_L^{th}). It was found that the experimental values of n_2 in toluene, nitrobenzene, bromobenzene, and CS_2 are in good agreement with values calculated from the optical Kerr constants (see Table II). In pure and mixed benzene the measured numbers of n_2 are somewhat larger than those predicted from the Kerr constants, indicating that in this substance the Kerr effect is probably not the only mechanism responsible for the intensity-dependent refractive index.

Using the known values of the Raman gain factors g_S and the measured values of n_2 we calculated the dimensionless gain factors $\bar{g}$ for the liquids (see Table II). In the mixtures of benzene and CS_2, SRS occurred with the frequency shift of the benzene vibration (here, frequency is measured in wave-number units 992 cm^{-1}) because of the small amount of CS_2 ($\leqslant 14\%$).

In Fig. 7 the experimental values of the limiting diameters of the laser light are plotted versus $\bar{g}$ for benzene (●), 93% benzene-7% CS_2[24] (▽), 86% benzene-14% CS_2 (▼), and toluene (○). The solid line has been calculated (see Sec. II A) using our model of forward SRS. In the calculations a $1/e$ radius at the entrance window of 350 μm was used, which was typical for these experiments. The agreement between the measured points and the calculated curve is satisfactory allowing for the limited accuracy of g_S and n_2 and the variations of d_L for different laser shots (vertical error bars). The experimental findings are readily explained by our model. Benzene (●) has a con-

TABLE II. Nonlinear refractive index n_2 obtained from Kerr constants (K) and from measured threshold power values (th), Raman gain factor g_S, normalized gain factor $\bar{g}$, and relaxation times of the molecular vibrations and the Kerr effect, τ_R and τ_K, respectively.

	$10^{12}n_2$[a] (cm^3/erg)		$10^3 g_S$		τ_R[b]	τ_K[c]
	K	th	(cm/MW)	$\bar{g}$	(psec)	(psec)
Benzene	1.5[d]	2.5	3.0[c]	2.5	2.3	1.6 3.8
Benzene-CS_2						
93%-7%	2.1[e]	2.9	2.9[f]	2.1		
86%-14%	2.7[e]	3.6	2.7[f]	1.6		
Nitrobenzene	10.4[g]	9.6	1.8[h]	0.4	0.8	48
Bromobenzene	5.6[g]	4.6	1.5[c]	0.7	2.8	15 11
Toluene	2.6[g]	2.5	1.4[h]	1.2	2.7	1.8 4.1
CS_2	11.9[i]	13	17[h]	2.9	10.6	2.0

[a] The nonlinear refractive index n_2 was calculated from the Kerr constant B and the wavelength λ of the light by $n_2 = \frac{2}{3}\lambda B$. The nonlinear dielectric constant is given by $\epsilon_2 = 2n_0 n_2$.

[b] Calculated from the Raman linewidth $\delta\bar{\nu}_R$ given in Ref. 26; $\tau_R = 1/2\pi c\delta\bar{\nu}_R$.

[c] Reference 27.

[d] Reference 28.

[e] Reference 29.

[f] The variations of the Raman cross section with concentration are taken from Ref. 30. The Raman linewidth is assumed to be the same as in pure Benzene.

[g] Reference 31.

[h] The values of g_S are calculated from the spontaneous scattering cross section given in Ref. 32 and the linewidth given in Ref. 26.

[i] Reference 33.

siderably larger diameter than toluene because of its higher Raman gain factor g_S. The drastic decrease of d_L in the mixtures of benzene and CS_2 (▽,▼) is mainly due to the increase of n_2 by the admixture of CS_2 to benzene (see Table II). It should be mentioned that the limiting diameters of nitrobenzene, bromobenzene, and pure CS_2 cannot be explained with the steady-state theory and require a discussion of the transient case.

2. *Transient situation*

Using the value of $\bar{g}=2.9$ for CS_2 (see Table II) we calculate a limiting diameter of $d_L \simeq 73$ μm from the steady-state theory which disagrees with the measured value of 6 μm. Agreement between theory and experiments is obtained for a smaller effective Raman gain factor of $g_S^{\text{eff}}=8.7\times10^{-3}$ cm/MW, corresponding to $\bar{g}=1.5$. For an explanation it should be recalled that in our experiments one focal spot moves with high velocity through the end of the liquid cell. The transit time was calculated in Sec. II B to be of the order of several tens of picoseconds. The dephasing time of the molecular vibrations in CS_2 is of the same order of magnitude ($\tau_R=10$ psec; see Table II). Therefore, transient Raman scattering occurs in the focal region with a reduced gain factor leading to a small value of the limiting diameter.

The following rough estimate shows that a reduction of the steady-state Raman gain coefficient by a factor of 2 is reasonable. The cell length is divided into two regions: the focal region with a length $\Delta l_f=0.07$ cm and the remaining part of the cell ($l\simeq 30$ cm) with intensities $I_f=24$ GW/cm^2 and $I_L=52$ MW/cm^2, respectively. These numbers are taken from the results of the computer calculations for CS_2. The transit time Δt_f of the focal spot in CS_2 is approximately 14 psec (see Sec. II B) which is of the order of the vibrational dephasing time. The transient Raman gain factor g_S^f in the focal region is at least smaller by a factor of 3 than the steady-state value g_S.[21] Assuming that in the remaining part of the cell stationary stimulated Raman scattering occurs, we estimate an effective gain factor g_S^{eff} from the relation

$$g_S^{\text{eff}}(I_L l + I_f \Delta l_f) = g_S I_L l + g_S^f I_f \Delta l_f \,.$$

Using the numbers given above, we obtain $g_S^{\text{eff}}=11\times10^{-3}$ cm/MW or less. This value should be compared with $g_S^{\text{eff}}=8.7\times10^{-3}$ cm/MW used in the computer calculations.

The assumption of transient stimulated Raman scattering in CS_2 in the focal region is further substantiated by experiments with laser pulses having a longer time duration. When the duration of the laser pulses was changed from 15 to 23 nsec, the limiting diameter in CS_2 increased from 6 to 20 μm. This is readily explained by the fact that for longer pulses SRS is near the quasi-steady-state with a large value of the minimum diameter.

In bromobenzene and nitrobenzene the nonlinear refractive index n_2 is larger than in most of the other liquids leading to small values of the normalized gain factor $\bar{g}=0.7$ and 0.4, respectively (see Table II). For these values of $\bar{g}$, a Raman-limited beam diameter of <1 μm is expected from the steady-state theory (see Fig. 7), while in our experiments approximately 15 μm have been measured (corresponding to $\bar{g}=1.9$). We try again to explain the results qualitatively by the rapid movement of the focus at the end of the liquid cell. In contrast to CS_2 the Raman effect should be nearly quasistationary while the orientational Kerr effect exhibits a transient behavior due to its long relaxation time (see Table II). In the short time the focal spot moves through a small volume element the nonlinear refractive index cannot reach its full steady-state value. A larger value of the limiting diameter occurs in this case, as discussed in Sec. II B. In this way better agreement between the calculated and measured minimum diameter is obtained.

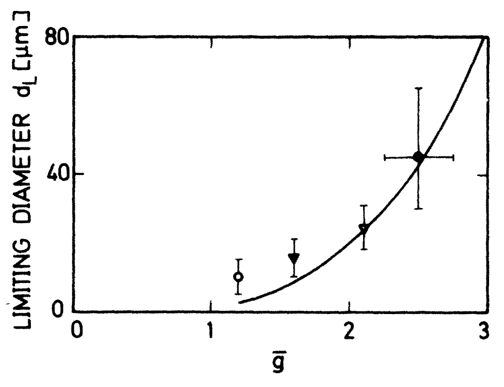

FIG. 7. Limiting diameter d_L of the laser light versus normalized gain factor $\bar{g}$. The symbols represent the experimental points: closed circle, benzene; open triangle, 93% benzene-7% CS_2 (Ref. 24); closed triangle, 86% benzene-14% CS_2; open circle, toluene. The solid line is calculated using the following parameters: $\bar{E}_L(0,0)=7.19$; $\bar{E}_S(0,0)=3\times10^{-6}$; $\bar{\kappa}=0.93$; $\bar{D}_V=1.0$; $a_L=350$ μm.

IV. SUMMARY

The origin of the limiting diameters of the self-focusing of laser light has been discussed in this paper under the following experimental conditions: The light beam of a single-transverse- and longitudinal-mode giant pulse laser with a pulse duration of the order of 10^{-8} sec is incident on a liquid in which the Kerr effect gives the main contribution to the nonlinear refractive index; the focal spot is moving with high velocity through the end of the liquid cell.

Computer solutions of the coupled differential equations for the laser light and the first Stokes line of *forward* stimulated Raman scattering showed that high conversion of laser light into forward Raman light limits the self-focused beam diameter. The dependence of the limiting diameter on nonlinear refractive index n_2, Raman gain factor g_S, incident laser power P_L, length l of the medium, beam divergence D_V, and initial conditions of the Raman light was calculated and discussed.

We measured the limiting diameters of several pure and mixed liquids. In benzene, mixtures of benzene and CS_2, and in toluene fair agreement was found between the experimental value of the limiting diameter and the size of the focal spot as estimated from the steady-state theory. In liquids where the relaxation time of the molecular vibrations or the molecular reorientation is long, the transient behavior of stimulated Raman scattering or self-focusing has to be taken into account because of the rapid movement of the focal spot through the far end of the liquid cell. In CS_2 the measured limiting diameter is smaller, in bromobenzene and nitrobenzene it is larger than that calculated from the steady-state theory. The experimental results in CS_2 have been qualitatively explained by the occurrence of transient stimulated Raman scattering due to the long relaxation time of the molecular vibrations in this liquid. In nitrobenzene and bromobenzene the long orientational relaxation time causes a transient behavior of the Kerr effect in the focal spot and is believed to be the reason for the large measured limiting diameters.

In conclusion, we believe that in experimental situations where high conversion of laser into forward Raman light occurs, the mechanism described in this paper plays an important role in limiting the self-focused beam diameters.

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$$I_S = I_S(0, 0)\ \exp[g_S I_L(0, 0) z e^{-r^2/a_L^2}],$$

the radius a_S of the Stokes light is readily calculated. For low conversion efficiency at the end of the medium [$g_S I_L(0, 0)z \simeq 20$ to 30], we obtain $a_S/a_L \simeq 1/[g_S I_L(0, 0)z]^{1/2}$, i.e., a_S is small compared to a_L. With increasing distance z the Raman beam diameter decreases continuously (outside the saturation region).

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