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A NEW CLASS OF TRAPPED LIGHT FILAMENTS*

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The existence of self-trapped filaments of light in intense laser beams and their influence on stimulated Raman radiation have already been discussed.[1, 2, 3] Furthermore, filaments of diameters about 50μ which are self-focussed by the Kerr effect have been identified and most of their properties are found to agree with expectations.[3, 4, 5] However, it is shown here that the stimulated Raman radiation is characteristically associated with a second class of filaments of much smaller diamter, of different threshold properties, and with characteristics which affect substantially the nature of Stokes and anti-Stokes radiation.

A typical photomicrograph of light produced by a Q-switched ruby laser just as it emerges from the end of a 50 cm cell of carbon disulfide is shown in Figure 1a. A beam from a 0.5 mm pinhole such as that used in reference 4 provided the exciting light. The Airy disk and Airy ring of the Fraunhofer diffraction from the pinhole form the background light in the figure. The bright central region is caused by self-trapping due to the Kerr effect, as observed previously. In addition, superimposed on this

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concentrated filament are many bright filaments of laser light of much smaller diameter, about 5 microns. We shall refer to the former as large-scale trapping and to the latter as small-scale trapping.

The Stokes light, a typical photomicrograph of which is shown in Figure 1b, occurs in small filaments of about the same size as the small-scale laser filaments. By reflections from a partially reflecting mirror in the laser beam inside a liquid cell, these small-scale filaments have been shown to persist for many centimeters. The technique was similar to that used previously for large-scale trapping,[4] but the mirror consisted of a dielectrically coated six micron thick mylar film rather than a thicker microscope slide in order not to destroy the filaments by allowing diffraction inside the glass slide. Characteristically, the many filaments of Stokes radiation which occurred at 19 cm inside the cell persisted to the end of the 25 cm cell in the same relative positions.

Photomicrographs of the beam as it emerged from cells of carbon disulfide of varying lengths show the following characteristics. At four centimeters path length no focussing effects or stimulated Raman radiation were observed in a diffraction-limited beam of 200 kwatts power and 500 microns diameter. The self-focussing distance[6] was approximately six centimeters. In cells of this length the concentration of light into large-scale trapping is very much in evidence. Figure 2a shows the beam as near self-focus as we were able to obtain and demonstrates a central bright filament about five microns in size. Immediately

upon the onset of large-scale trapping, bright filaments of Stokes radiation approximately five microns in size appeared. Associated with such filaments was a marked decrease in the intensity of laser light at the position of these filaments. This decrease can be larger than an order of magnitude and shows up as dark spots the order of five microns in size within the large-scale self-focussed region, as shown in Figure 2b. By 10 cm path length, the laser beam was found to contain bright filaments rather than dark spots, which occurred in the same positions as the bright Stokes filaments and were also approximately five microns in size. Bright filaments about the same size were observed at twenty-five and fifty centimeters cell length (cf. Figure 1). By one meter cell length these filaments no longer appeared prominently. Similar small-scale filaments were observed in the other Raman-active liquids we investigated: nitrobenzene, bromobenzene and toluene.

While near the large-scale focussing distance the Stokes intensity increased very rapidly, in succeeding paths up to fifty centimeters it increased in intensity by less than two orders of magnitude. Near the focussing distance large-scale trapping was very rarely observed without intense Stokes filaments and their associated dark laser spots. Hence the region of high gain for the Stokes light must be less than a few millimeters, with a gain as large as about e^{30} per centimeter. This rapid conversion weakens the laser intensity within the first few millimeters, producing the observed dark spots. The majority of anti-Stokes

light radiated in cones was observed to be emitted in this high-gain region.

The thresholds for stimulated Raman emission and small-scale trapping appear to be the same as for large-scale focussing. Near the self-focussing distance, stimulated Raman filaments were always observed in conjunction with large-scale focussing. Transient self-focussing may occur at power levels lower than those resulting in steady-state trapping.[7] In this case small-scale filaments were formed which remained trapped for long distances even when the large-scale focussing was no longer apparent. While there were no small-scale filaments at power levels below those necessary for self-focussing, a small increase of incident power above threshold led to many filaments, in striking contrast to the axially symmetric large-scale intensity distribution.

The total energy transmitted through each small-scale filament was measured photographically at the end of twenty-five centimeters of liquid and found to be a few ergs. Both the laser and Stokes filaments appear to last approximately one nanosecond rather than the full 30 nanoseconds duration of the laser pulse.[2, 8] Thus the total power in each filament was few tenths to one kilowatt, which is an order of magnitude or more below that required by theory for either Kerr or electrostrictive effects to produce trapping. The change in index of refraction required to trap light in a filament of diameter d is given by $\Delta n = \frac{n}{2}\left(\frac{.61\lambda}{nd}\right)^2$. For the five micron filaments in carbon disulfide, this gives an order of magnitude or more larger than can be obtained from known nonlinearities due to Kerr or electrostrictive effects in the optical fields which must be present.

The close relation of small-scale trapping to the stimulated Raman effect suggests an increase in the refractive index due to molecular excitation. This can arise from the increased molecular polarizability of excited vibrational states. While accurate numbers are not known, the magnitude of this effect can be estimated from the rotation-vibration constants. These show that molecules typically change moment of inertia by a good fraction of one per cent due to vibrational excitation, and hence their average dimensions expand by this amount. Since the polarizability is roughly proportional to the cube of the molecular dimension, the molecular expansion due to excitation would then increase the index of refraction of the material by an amount comparable with one per cent if all the molecules are excited. Excitation of about ten per cent of the molecules will lead to refractive index changes consistent with those observed in small-scale trapping. Except in the region of very high Stokes gain, production of the required degree of excitation appears somewhat marginal from the present experimental numbers, even when one takes into consideration that a single laser photon may be completely converted into vibrational energy by successive Stokes emission and subsequent direct infrared absorption, and that the total energy absorbed in the filament may be greater than that which is transmitted.

None of the other mechanisms we have investigated appear to give a large enough refractive index change in the presence of the experimental field strengths. Higher order terms in the

simple Kerr or electrostrictive effects are expected to be small and of the wrong sign to produce an increased change of index of refraction of the observed type. A cross-term between Kerr and electrostrictive effects could at least have the correct sign, but it appears much too small to be effective in these cases. One might wonder whether some change in phase, such as crystallization induced by molecular alignment and pressure of the field, could occur. The energy in the molecular polarization, however, is very much less than kT, and we have found experimentally that the properties of these filaments in mixtures are a continuous function of relative concentration, which indicates that no phase transition is present.

It is not difficult to understand the short time duration of the filaments. The vibrational energy given to the molecules through the stimulated Raman effect will be converted after a characteristic relaxation time into translational energy, causing the filament to heat up and expand. Expansion will decrease the refractive index and destroy the trapping. In CS_2 the characteristic time for vibrational relaxation to translational energy is 2×10^{-9} sec.[9] The characteristic time for destruction of a filament by expansion is the time required for sound to move a sizeable portion of the filament diameter, also approximately one nanosecond. Thus the duration of small-scale trapping for only a few nanoseconds is not surprising. Furthermore, it is clear that the decrease of refractive index due to material expansion would keep radiation out of this region once the light filament has been destroyed. Thus the appearance of dark spots in the laser distribution near the region of high Raman gain may persist the entire duration of the laser pulse even when the Stokes radiation lasts only one nanosecond.

The cause of the low Raman gain in the latter portions of long cells has not been worked out in detail. While saturation of the molecular population difference is an attractive mechanism, the present experimental numbers do not appear to allow a sufficient fraction of the molecules to be excited. Decrease in gain because of the presence of many orders of Stokes radiation is certainly a contributing factor, but does not adequately decrease the conversion of laser light since the highest order Stokes radiation is absorbed directly by the liquid and provides a rapid loss. Dispersion in the refractive index will cause further gain decrease if the exciting light is broadened in frequency.

The existence of small-scale trapping described here should have a considerable effect on the characteristics of stimulated radiation through the following new considerations:

(a) The small diameter of the filaments changes the phase velocity of light waves by wave-guide effects and hence modifies the angles of anti-Stokes radiation.[10]

(b) The large change in index of refraction also modifies phase velocities and radiation angles.[10]

(c) The large change in index is probably accompanied by an appreciable change in acoustic velocities.

(d) The index change in the filaments produces more powerful modulation effects and smearing of radiation frequencies than what is calculated assuming Kerr effects alone.[11]

(e) The rate of change of index of refraction and hence of phase of the trapped radiation associated with rapid formation

and decay of the small-scale filaments can contribute importantly to frequency smearing.[10]

(f) Variations in molecular saturation and in index of refraction along the filament can give different relative intensities of Stokes radiation in the backward and forward directions, as is sometimes observed.

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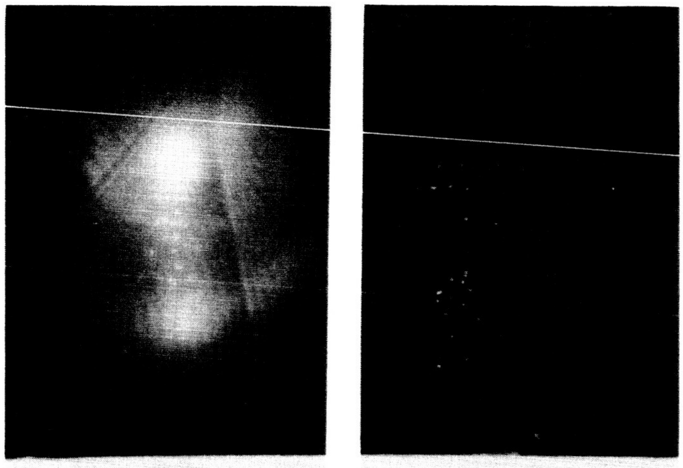
Figure 1

(a) Image of a ruby laser beam emerging from a 50 cm cell of CS_2 and exhibiting large and small-scale trapping. Magnification is 30x. The bright central portion is the large-scale trapped beam; the many small bright filaments demonstrate the small-scale trapping. The broad disk and ring of light are the untrapped beam diffracting from the initial pinhole.

(b) Stokes radiation under conditions similar to (a). Magnification 50x.

Figure 2

Image of ruby laser beam emerging from a 6 cm cell of CS_2 under magnification a factor of 40x. (a) Very near self-focus (b) Slightly higher power level, i.e., self-focus occurred just before the end of the cell.



a b

FIGURE 1

a b

FIGURE 2