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INTENSITY-INDUCED OPTICAL ABSORPTION CROSS SECTION IN CS_2

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This Letter reports the direct observation of intense radiation-induced absorption in liquid carbon disulfide and in several other "highly transparent" materials. This effect was first observed during an attempt to stimulate Raman laser emission[1,2] in CS_2. The induced absorption produced a strong cavity loading which apparently precluded Raman laser emission. To study this effect, we have made direct absorption measurements at 6943 Å. For CS_2 we find a total absorption cross section σ linear in incident photon flux F, $\sigma = \sigma_0 + \sigma_1 F$, with $\sigma_1 = (5 \pm 4) \times 10^{-51}$ cm^4 sec.

Absorption measurements were made using the external beam of a Q-switched[3] ruby laser. The parallel, linearly polarized beam had the following characteristics: peak power, 1.0×10^6 watts; pulse duration, 40 nsec; spectral width of emission at 6943 Å, 2 cm^{-1}; and angular diameter at half-intensity, 0.24°.

To enhance the nonlinear absorption, the beam was focused within the sample by a lens having a 4.83-cm focal length in air. Portions of the incident and transmitted beams were reflected by 45° glass plates onto diffusely scattering white surfaces viewed through a distant lens by matched 925 phototubes provided with 6940 ± 100Å interference filters. The phototube signals were displayed on a dual-beam oscilloscope having a 12-nsec rise time.

Incident beam intensity was varied over a 20-dB range by appropriate reflecting attenuators; the laser power level was held fixed to avoid any systematic variation of the complex intensity distribution near the focus. Similarly, possible gain-bandpass errors were minimized by holding the detector signals approximately constant through the use of calibrated attenuating filters in front of the detector lens.

Figure 1(a) shows the dependence of transmitted peak power, P_t, on indicent peak power, P_0, observed for spectroscopic grade CS_2 at room temperature. The probable error is estimated to be ±30% in the absolute P_0 scale, and ±20% in the P_0/P_t scale. The data were taken over a five-week period and showed no systematic trend

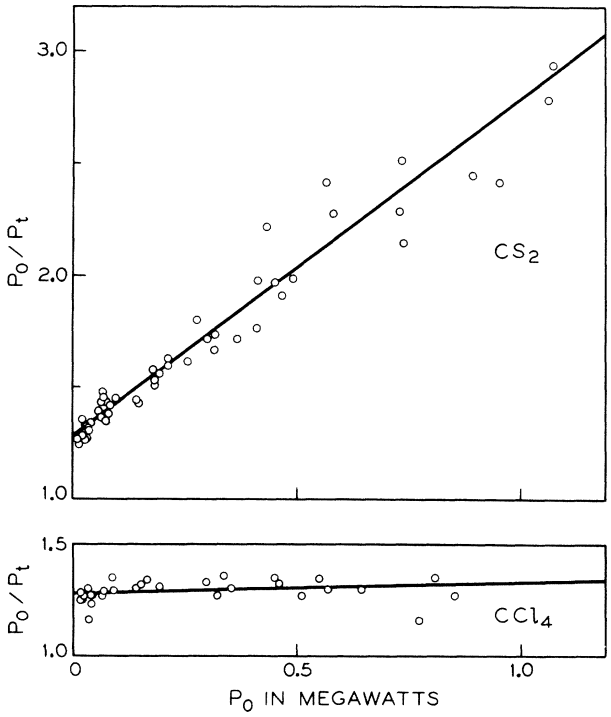

FIG. 1. Ratio of incident power P_0 to transmitted power P_t as a function of P_0.

as various laser components deteriorated with use.

The linear absorption coefficient at 6943 Å was measured by conventional techniques and found to have the value $(2.0\pm0.3)\times10^{-3}$ cm^{-1} and to be independent of wavelength in this region. Since this value includes losses from scattering, etc., it sets an upper limit to the linear absorption cross section of $\sigma_0 \leq 2.0\times10^{-25}$ cm^2.

To analyze the induced absorption, consider an effective cross section per molecule $\sigma=\sigma_0+\sigma_1 F$, and assume that $\sigma_1 F \gg \sigma_0$ holds. Then, the decrease in beam power P in the beam direction z is $dP/dz = -N\sigma_1 P^2/[h\nu A(z)]$, where A is the beam area and N is the molecular density. If it is further assumed that the intensity distribution over $A(z)$ is uniform, then $P_0/P_t = 1+[N\sigma_1 P_0/(h\nu)]\int dz/A(z)$. To an excellent approximation, the integral $\int dz/A$ is $8\,\mathfrak{F}/[\pi d]$, where $\mathfrak{F}$ is the $f/$ number of the convergent beam and d is the focal spot diameter. From Fig. 1(a) and the measured values $d=2.0\times10^{-2}$ cm, $\mathfrak{F}=2.8\times10^{1}$, $N=1.00\times10^{22}$ cm^{-3}, and $\nu=4.32\times10^{14}$ sec^{-1}; we obtain a value for σ_1 of 9.5×10^{-51} cm^4 sec. Since this value is obtained under the assumption of uniform intensity distribution, it represents an upper limit to the true value.

Examination of the angular distribution of the maser emission allows us to infer the maximum local flux and permits us to set a lower limit to σ_1. Our observations showed that the beam diameter at half-maximum intensity was $d_0=4.2\times10^{-3}$ rad, that the emission consisted of a random, overlapping array of roughly circular lobes of diameter $d_1=(1.9\pm0.4)\times10^{-4}$ rad,[4] and that these intensity variations disappeared when examined with a resolution $d_2=5\times10^{-4}$ rad. We conclude that the local flux could have exceeded average flux by a maximum value of $(d_2/d_1)^2\sim7$, and that a lower limit to σ_1 is thus 1.4×10^{-51} cm^4-sec. Our best estimate of σ_1 is the average value $\sigma_1=(5\pm4)\times10^{-51}$ cm^4 sec.

For incident peak powers in excess of 5×10^5 watts, intense off-axis scattering rendered the focus visible as a brilliant red filament. The irregular appearance of this region suggested that bubbles or other optical inhomogeneities produced at the focus were responsible for the scattering. The intensity of the light scattered at 90° to the beam axis was measured both parallel and perpendicular to the incident $\vec{E}$ vector with the aid of a 925 phototube provided with a 6940±100Å filter. The perpendicular light was almost completely polarized parallel to $\vec{E}$ and was about four times as intense as the parallel light, which was not linearly polarized. The scattered intensity varied about 50% from pulse to pulse, but on the average was proportional to P_0^n with n in the range 1.5-1.9. Using the measured perpendicular intensity at $P_0=1\times10^6$ watts and assuming isotropic scattering, we calculate a maximum scattering loss of $0.05P_0$. We conclude that scattering from optical inhomogeneities does not make a significant contribution to the observed σ_1. Further, we found that 90° scattered light at all other wavelengths in the range 6000-9000 Å amounted to less than one-fifth that in the 6943Å region.

The σ_0 losses are inadequate to produce significant local heating at the beam focus. With $P_0=10^6$ watts, they would generate a maximum temperature rise of 2°C. In contrast, the much larger σ_1 losses are compatible with local boiling.

Intense forward scattering was observed at the discrete frequencies 13 087 cm^{-1}, 13 742 cm^{-1}, 15 054 cm^{-1}, and 15 709 cm^{-1}. These differ from the observed laser frequency $\nu_0=14\,397.5$ cm^{-1} by integral multiples of $\nu_m=655$ cm^{-1}, the frequency of the CS_2 symmetric stretching vibration.[5,6] Light at these frequencies was found to be sharply peaked along the z axis. A 1.1° angular diameter at half-intensity was observed in the liquid for the most intense 13 742-cm^{-1} emission (angular diameter of laser beam 2.0°). This emission is similar to that found by Terhune[7] in various Raman-active materials and attributed to the processes of the type $2h\nu_0 \rightarrow h(\nu_0+\nu_m)$, in which the condition $2\vec{k}_{\nu_0}=\vec{k}_{\nu_0-\nu_m}+\vec{k}_{\nu_0+\nu_m}$ is satisfied. However, for our conditions no angular structure associated with the $\vec{k}$-vector constraint was observed or to be expected, the estimated ring diameter of 0.3° being smaller than that of the laser beam.

We have measured the forward scattered peak light power at 13 742 cm^{-1} and find $P\propto P_0^n$, where $n\geq3$; for the 13 087-cm^{-1} line we find $n\geq6$. Photographic evidence indicates similar P_0 dependence for the anti-Stokes lines, each of which has an intensity less than, but comparable to, the corresponding Stokes line.

For incident power $P_0=1.0\times10^6$ watts, we find peak values $P(13\,742)=(2.8\pm1.0)\times10^5$ watts and $P(13\,087)=(5.8\pm2.0)\times10^4$ watts. The duration of this emission was less than 20 nsec. Since we observe insignificant forward Raman scattering for incident powers less than 0.5×10^6 watts, and since this process shows a strong dependence on P_0, we conclude that Raman scattering does not make an important contribution to σ_1 loss for

our range of input powers.

A variety of other materials transparent at 6943 Å have also been examined for nonlinear loss at $P_0 \approx 0.7\times10^6$ watts. These are listed below in order of decreasing fractional absorption $K=(P_0-P_t)/P_0$: $0.6 \geq K \geq 0.4$, nitrobenzene, benzene, carbon disulfide; $0.3 \geq K \geq 0.1$, bromobenzene, pyridine, polymethylmethacrylate, iodobenzene, acetone, chlorobenzene, chloroform, acetophenone; $0.1 \geq K$, trichloroethylene, cyclohexane, methanol, toluene, carbon tetrachloride, fused quartz, water, isopentane, 2,2,4-trimethylpentane, nitromethane. Figure 1(b) shows a typical power dependence for a material in the last group.

Not all the nonlinear absorbers show the relation $\sigma \propto F$. Benzene, for example, shows negligible absorption at $P_0 = 0.5\times10^6$ watts and has relatively little loading effect when placed in a laser resonator. However, at higher incident powers absorption rises rapidly, and is accompanied by intense Stokes and anti-Stokes Raman emission.

The experimental results appear to rule out inhomogeneity and Raman scattering as the source of σ_1 loss in CS_2. This loss, linear in F, is consistent with a process in which two photons are simultaneously absorbed and a CS_2 molecule is excited to the $^1B_2$ state.[8] A theoretical description of such processes has been given,[9] and we may estimate σ_1 for two-photon absorption from Kleinman's[10] formula $\sigma_1 = r^2c^2f^2/n^2\nu_0^2\Delta\nu$, where n is the refractive index, r is the classical electron radius, and $\Delta\nu$ is the width of the real, excited state at $2h\nu_0$. In the derivation of this expression it is assumed that the absorption occurs through a single intermediate state coupled to both the initial and final states by an oscillator strength f. Using this formula with $\Delta\nu = 1\times10^{14}$ sec^{-1}, $\nu_0 = 4.32\times10^{14}$ sec^{-1}, and $f=1$, we obtain $\sigma_1 = 1.5\times10^{-48}$ cm^4 sec. The observed σ_1 corresponds to an effective f value of 0.1 and is fully consistent with the two-photon absorption process. In order to corroborate the two-photon mechanism, it would be of interest to extend this experiment to wavelengths greater than 7800 Å, since there is negligible absorption in CS_2 at wavelengths longer than 3900 Å.

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HYPERFINE STRUCTURES OF THE RESONANCE LINE OF THE ARC SPECTRA OF THE ISOTOPES 135 AND 137 OF BARIUM

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The hyperfine structures of the line Ba I, 5535 Å ($6s^2\,^1S_0 - 6s6p\,^1P_1$), of the isotopes 135 and 137 of barium are of particular interest because they enable the quadrupole moment coupling factor, B, to be calculated for the level $6s6p\,^1P_1$. Approximate values have been found by the present authors[1] from an earlier measurement of the structure of the line given by natural barium combined with measurements of the isotope shifts and the already known ratios, for the two isotopes, of the magnetic-moment splitting factors, A, and the quadrupole-moment coupling factors, B. But the values obtained were lacking in precision on account of some uncertainty in the values of the