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INTENSITY-DEPENDENT CHANGES IN THE REFRACTIVE INDEX OF LIQUIDS

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In this paper we wish to report measurements of some of the changes in the real part of the index of refraction for monochromatic light which are proportional to the light intensity. In the accompanying analysis of the effect, for elliptically polarized light in isotropic centrosymmetric materials, it is shown that in addition to a polarization-independent change in velocity, a rotation of the axes of the vibrational ellipse as a function of distance occurs. Observation of this rotation, such as reported here, provides a very sensitive technique for measuring the magnitude and sign of the coefficients involved. This rotational effect is not to be confused with the Faraday effect or optical activity. It is more closely related to the second-order Kerr and Cotton-Mouton effects, which involve dc fields, and are special cases of the interaction between two frequency components. Such an interaction, however, leads in isotropic centrosymmetric materials to birefringent rather than rotary effects.

The effect which usually determines the intensity-dependent index of refraction can be described in terms of an induced nonlinear polarization of third order in the electric field strength.[1] The Fourier component of this nonlinear polarization at frequency ω, $\mathrm{Re}\vec{P}^{\mathrm{NL}}$, which is induced by a plane wave at frequency ω traveling in the z direction can be represented as follows[2]:

$$P_i^{\mathrm{NL}} = [AE_i(\vec{E}^*\cdot\vec{E}) + \tfrac{1}{2}BE_i^*(\vec{E}\cdot\vec{E})] \times \exp(-i\omega t + in\omega z/c), \quad (1)$$

where the electric vector of the light wave is $\vec{\epsilon}(\omega,z) \equiv \mathrm{Re}[\vec{E}\exp(-i\omega t + in\omega z/c)]$, n is the refractive index at frequency ω, and A and B are complex constants. Here, $\vec{E}$ is regarded as a slowly varying function of z as a result of the nonlinearities. In a lossless medium, A and B are real and when ω is far from all resonances, $A \cong B$. Transforming $\vec{P}^{\mathrm{NL}}$ and $\vec{E}$ to a circular representation with $E_+ \equiv (E_x + iE_y)/\sqrt{2}$ and $E_- \equiv (E_x - iE_y)/\sqrt{2}$, it can be shown that in a lossless medium, the nonlinearity leads only to the following changes in the refractive indices for the two senses of circular polarization:

$$\delta n_+ = (2\pi/n)[AE_+E_+^* + (A+B)E_-E_-^*],$$

$$\delta n_- = (2\pi/n)[AE_-E_-^* + (A+B)E_+E_+^*]. \quad (2)$$

From the above, one can readily verify that the index changes for plane and for circularly polarized light are different. Let α be defined as the angle of inclination of the vibrational ellipse of the elliptically polarized radiation, measured from the x toward the $+y$ axis. Then α is equal to one half the phase difference between E_+ and E_- and varies with z as follows:

$$\alpha = \alpha_0 + \tfrac{1}{2}(\omega/c)(\delta n_+ - \delta n_-)z = \alpha_0 + (\pi\omega/cn)B(E_-E_-^* - E_+E_+^*)z. \quad (3)$$

The direction of rotation is determined by the sign of B and the handedness of the ellipticity.

Figure 1 is a schematic diagram of the experimental arrangement used. A giant pulsed ruby laser providing in the sample a 16-mJ pulse with a 40-nsec halfwidth was used. The mica eighth-wave plate was oriented so that the electric vector of the laser beam bisected the angle between its fast and slow axes. The laser beam was brought to a focus in the middle of a one-meter-long liquid cell with windows selected for minimum birefringence. A right-angle Rochon prism separated the output beam into two components plane-polarized parallel to the fast and

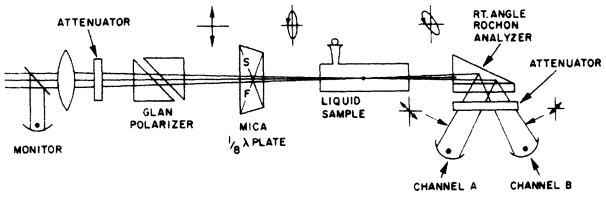

FIG. 1. Experimental arrangement used for detecting intensity-dependent rotation of the vibrational ellipse. The polarization of the beam as seen when facing into the laser is indicated at several positions.

to the slow axes of the eighth-wave plate. The two channels received equal power until the balance was destroyed by rotation of the vibrational ellipse. The intensity of the laser beam at the sample was varied by moving Corning filters from the front to the rear attenuator. The experimental arrangement was designed to minimize effects due to birefringence in the lens and filters. By also using the monitor, intensity-dependent losses[3] could be detected. They were found to be negligible except as noted. In addition, all measurements were made at power levels below the observed threshold for Raman laser action.

Typical results are shown in Fig. 2 for bromoform. The upper data were obtained with the slow axis of the eighth-wave plate along direction A. The lower data were obtained when the mica was rotated 90°. Thus, experimentally, as the intensity is increased, the vibrational ellipse rotated as a whole in the same direction as the electric vector at a fixed point in space. A maximum rotation of approximately sixteen degrees is indicated. All effects vanished when the ellipticity was reduced to zero, and also when the sample was removed. At low power levels the

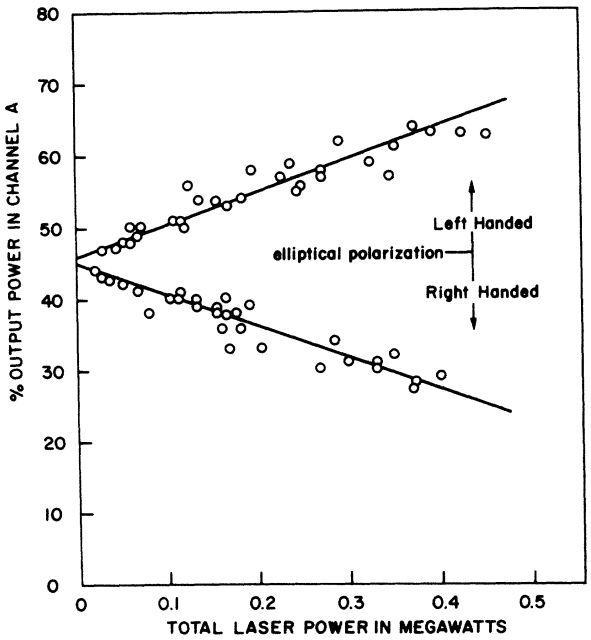

FIG. 2. Measured percentage of total power at the output polarized in direction A vs laser power with bromoform as the sample. Upper data with slow axis of eighth-wave plate in direction A. Lower data with eighth-wave plate rotated 90°. Data points indicate results for individual laser pulses.

percentage of the total energy in channel A departed slightly from 50% because of residual birefringence and slight misalignment of the optical components. These also caused a slight difference of slope and intercept for the two lines fitted to the data in Fig. 2. The increased scatter in the data when the nonlinear effects are large is presumably due to changes in the time and spatial structure of the laser beam from pulse to pulse.

Table I summarizes data taken in a range of

Table I. Measured amounts of intensity-dependent rotation and inferred nonlinear susceptibility constants in various liquids ($\alpha \pm 1°$).

Liquid	P_T (megawatts)	α (degrees)	$B \times 10^{15}$ (cm^3 erg^{-1})
Water	0.4[a]	1	2
Methanol	0.4	1	2
n-Hexane	0.4	2.5	5
Carbon tetrachloride	0.4[b]	3.2	7
Chloroform	0.25[b]	9	30
Bromoform	0.35[b]	16	40
Pyridine	0.15[b]	10	60
Carbon disulfide	0.05[b]	13	250

[a]Threshold for Raman laser action $\cong 3 \times 10^6$ watts.
[b]Threshold for Raman laser action.

liquids. The angle α was deduced from the upper data of Fig. 2 using the relation $P_A/P_T = (1+0.707 \times \sin\alpha)/2$, where P_A is the power in channel A and $P_T = P_A + P_B$ is the total laser power. To compute B from α, the relation $\alpha = (10\omega^2/nc^3)P_T B$ was used. This equation was derived from (3) by assuming that the entire rotation takes place in an effective cylindrical focal volume whose length to cross-sectional area ratio is $1.2n/\lambda$, within which the beam has uniform intensity. This model is based upon the intensity distribution in the region of a diffraction-limited focus.[4] The ratio has been degraded by a factor of 3, which represents a best guess as to the effects of the observed departures from an ideal focus. Also the pulse shape was assumed square in time with duration equal to the measured half-width. The values thus deduced for B cover the same range as those obtained for similar materials from three-wave frequency-mixing experiments carried out away from a focus,[2] and third-harmonic generation experiments.[5]

In all of the liquids studied B was found to be positive. A is expected to have the same sign as B.[2] Both constants being positive implies a decrease in velocity and an increase in Rayleigh scattering with increasing intensity. In a liquid with $A \cong B \cong 10^{-14}$, changes in the refractive index of the order of $1:10^3$ will occur at an $f/10$ ideal focus of a one-megawatt laser beam. This intensity-dependent slowing should be considered in the calculation of the angles at which anti-Stokes radiation will emerge[5,6] from such a focus when Raman laser action occurs.

The high value of B for carbon disulfide arises from the two-photon absorption resonance reported by Giordmaine.[3] Its positive sign implies that 2ω is less than the resonance frequency. His value for the intensity-induced scattering cross section for plane polarized light gives $\mathrm{Im}(A + \frac{1}{2}B) = (5 \pm 4) \times 10^{-14}$ cm^3 erg^{-1}. We also have observed an intensity-dependent attenuation of 10% in carbon disulfide with $P_T = 5 \times 10^4$ watts. Using the same model for the interaction volume as in the calculation of ReB, we calculate $\mathrm{Im}(A + \frac{1}{4}B) = 2 \times 10^{-14}$ cm^3 erg^{-1}. We plan to simultaneously study the polarization dependence of the intensity-induced absorption and the intensity-induced rotation. From these data we will be able to obtain accurate relative values for ImA, ImB, and ReB.

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SLOW NEUTRON-DEUTERON CAPTURE AND THE STRUCTURE OF ^{3}H AND ^{3}He†

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Recent experiments on the elastic scattering of high-energy electrons from ^{3}H and ^{3}He show that the magnetic moment form factors for the two nuclei and the electric charge form factor for ^{3}H are all quite similar to each other, while the charge form factor for ^{3}He falls off somewhat more rapidly with increasing q^2 than do the other three.[1,2] These observations have been interpreted[2] as indicating that the like pair of nucleons (protons in ^{3}He and neutrons in ^{3}H) have a somewhat different spatial distribution than the odd nucleon; the form factors (Fourier transforms) of these probability distributions are called F_L and F_O, respectively. Since the charge in ^{3}He is carried primarily by the like pair, while the charge in ^{3}H and the moment in both nuclei are carried primarily by the odd nucleon, such a model is in at least qualitative accord with the observations.

A subsequent paper,[3] which makes use of the isotopic spin formalism, ascribes the difference between F_L and F_O mainly to interference between the dominant, fully space-symmetric $^2S_{1/2}$ state with $I = \frac{1}{2}$ (denoted by S), and a $^2S_{1/2}$ state of mixed symmetry that also has $I = \frac{1}{2}$ (denoted by S').[4]