$$\chi_{111}(2\omega, \omega, \omega) = \frac{-N_0 \frac{e^3}{m^2} V}{(\omega_0^2 - \omega^2 - i\Gamma\omega)^2 (\omega_0^2 - 4\omega^2 - i\Gamma 2\omega)} .$$

The approach of the fundamental frequency or of a harmonic to the natural frequency of the substance leads to an increase of the nonlinear susceptibility. Such an effect is observed, in particular, in semiconductors [5]. It is possible that the large nonlinear susceptibilities of the dyes are due to the proximity of the neodymium-laser second harmonic to the frequencies of the triplet-singlet transitions of the investigated compounds.

[1] N. Bloembergen, Amer. J. Phys. 35, 389 (1967).
[2] L.D. Derkacheva and A.I. Krymova, ZhETF Pis. Red. 9, 564 (1969) [JETP Lett. 9, 343 (1969)].
[3] M. Bass, D. Bua, R. Mozzi, and R.R. Monchamp, Appl. Phys. Lett. 15, 393 (1969).
[4] N. Bloembergen, Nonlinear Optics, Benjamin, 1965.
[5] R.A. Soref and H.W. Moos, J. Appl. Phys. 35, 2152 (1964).

STRUCTURE OF DEPOLARIZED COMPONENT OF SCATTERED LIGHT IN α-CHLORONAPHTHALENE

L.A. Zubkov, N.B. Rozhdestvenskaya and A.S. Khromov
Leningrad State University
Submitted 18 March 1970
ZhETF Pis. Red. 11, No. 10, 473 - 474 (20 May 1970)

The temperature dependence of the frequency of the transverse doublet observed in the depolarized component of scattered light [1 - 3] cannot be explained within the framework of the Leontovich phenomenological theory [4]. It is therefore of interest to measure the anisotropy relaxation time and the viscosity as functions of the temperature.

The measurements were made on α-chloronaphthalene, in which a depolarized doublet was distinctly observed and its behavior with changing temperature was investigated [3].

According to the Leontovich theory, the anisotropy relaxation time should be calculated from the half-width of the depolarized $I_{xy}(\omega)$ component of the scattered light. For many liquids, this component can be represented by a dispersion curve. The measurement setup was described in [3].

We have noted in the study of the $I_{xy}(\omega)$ component of scattered light in α-chloronaphthalene that a strong deviation from the dispersion curve with a single time τ is observed already in the region where the intensity decreases by a factor 10 - 15. The contour of the $I_{xy}(\omega)$ component can be described by a sum of two dispersion contours, one narrow with a sharp maximum and the other broad with a flat maximum; the half-widths of the contours are determined respectively by relaxation times τ_{cg} and τ_1. The relaxation time τ_{cg} was determined from the half-width of the narrow contour at height $I/2$. τ_1 was determined from the slope of a plot of $1/I = f(\Delta\omega^2)$. It should be noted that in this case I is the intensity in that frequency region $\Delta\omega$ where the contribution of the intensity from the narrow contour does not exceed 5%. In measurements of this type it is necessary to be certain that correct allowance has been made for the intensities due to the different orders of the interference. All the measurements were therefore made in the dispersion regions of 5 and 16.6 cm^{-1}.

$T°C$	$\eta \cdot 10^2$ poise	$r_1 \cdot 10^{12}$ sec	$r_2 \cdot 10^{12}$ sec
5	5.17	4.5	265
15	3.91	–	170
25	3.04	4.0	110
35	2.43	–	75
45	1.97	3.5	55

The measurements of τ_{cg}, τ_1, and η were made in the temperature interval from 5 to 45°C. As seen from the table, τ_{cg} and η change in this temperature interval by 5 and 2.5 times, respectively, while τ_1 changes insignificantly with temperature. Two relaxation times were first observed in nitrobenzene [5]. The presence of two relaxation times in a number of substituted benzenes has been pointed out in [6], but unfortunately the authors did not note the polarization of the scattered-light component, yet the measured value of τ depends strongly on the polarization. The measurements were made at one temperature.

The experimental data obtained by us are in good agreement with the Volterra theory [7]. According to Volterra, it is necessary to take into account in the theory of anisotropic scattering of light in a liquid at least two relaxation processes, namely the reorientation of the anisotropic molecules in the state of isotropic distribution of their axes (τ_1), and the relaxation process connected with a new distribution and with a possible displacement of the centers of the molecules (τ_2). Following Volterra, we can represent the $I_{xy}(\omega)$ component as a sum of two dispersion contours with half-widths corresponding to τ_1 and τ_{cg}, with τ_{cg} determined from the half-width of the narrow contour and connected with the time τ_2 by the relation $\tau_{cg} = \tau_2(1 + D)$, where D is the ratio of the integral intensities of the narrow and broad contours. The shear modulus is connected in the Volterra theory with the relaxation time τ_2 and is assumed to be independent of the temperature. Then, taking into consideration the expression for the frequency of the transverse doublet,

$$CO = q\sqrt{h/r_2(1 + D)} \; ,$$

(where q is the wave vector), we conclude on the basis of our experimental data that in the indicated temperature interval the factor (1 + D) decreases by one-half with increasing temperature, in good agreement with the theory under consideration. It should be noted that the previously measured 1.6-fold increase of the frequency of the transverse doublet with increasing temperature coincides in the given temperature interval [3], within the limits of experimental error, with the 1.4-fold increase calculated from formula (1).

[1] G.J. Stegeman and B.P. Stoicheff, Phys. Rev. Lett. 21, 202 (1968).
[2] I.L. Fabelinski, L.M. Sobirov, and V.S. Starunov, Phys. Lett. 29A, 414 (1969).
[3] N.B. Rozhdestvenskaya and L.A. Zubkov, Opt. Spektrosk., March 1970.
[4] M.A. Leoutovich, J. Phys. USSR 4, 499 (1941).
[5] A.K. Atakhodzhaev, M.F. Vuks, and V.L. Litivnov, Proc. 10th All-union Conference on Spectroscopy.
[6] H.G. Craddock, D.A. Jackson, and J.G. Powles, Mod. Phys. 14, 373 (1968).
[7] V. Volterra, Phys. Rev. 180, 156 (1969).