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Temperature dependence of the refractive index in condensed media

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A method of direct measurement of the classical constant of physical optics $(\partial n/\partial T)_\rho$ is proposed and developed. The essential element of the experimental set-up, which makes such a technique possible, is a single-mode laser with a pulse width of 3×10^{-9} sec and energy 0.3 J. A detailed substantiation of the proposed method is presented. Values of $(\partial n/\partial T)\rho$ were measured for a number of substances. The results are discussed and compared with the results of calculations carried out by Raman.

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Investigation of the temperature dependence of the refractive index is one of the fundamental tasks of physical optics. Up to now the solution of this problem was restricted to the measurement of the so-called total derivative dn/dT of the refractive index with respect to temperature. It is generally customary to express the dependence of the refractive index on the various thermodynamic parameters by derivatives with respect to these parameters. In this traditional manner the temperature dependence of the refractive index $n(T)$ can be represented in the general case in the form

$$n(T)-n_0=\int\left\{-\alpha(T)\rho\left(\frac{\partial n}{\partial\rho}\right)_T+\left(\frac{\partial n}{\partial T}\right)_\rho\right\}dT, \qquad (1)$$

where n_0 is the refractive index at a certain fixed temperature, $\alpha(T)$ the coefficient of thermal expansion, and ρ the density. The sum under the integral sign is the total derivative dn/dT. The first term of this sum is due to the effect of thermal expansion, and the second to the temperature dependence of molecular polarizability.[1]

Knowledge of the total derivative dn/dT alone is often insufficient. Thus, for instance, if one of the terms in the expression under the integral sign in (1) is not known in addition to dn/dT, it is impossible to establish a correspondence between the quantity Δn measured experimentally and the temperature change ΔT in the case when ΔT depends on a space coordinate. In fact, thermoelastic stresses arise in this case and the thermal expansion coefficient, with fringe effects disregarded, must be replaced by the quantity

$$\alpha_{\text{eff}}\approx\alpha\frac{1+\nu}{3(1-\nu)}, \qquad (2)$$

where ν is Poisson's ratio. It is then impossible to determine ΔT from the measured Δn and the known dn/dT.

In recent years the study of the temperature dependence of the refractive index has become a problem of timely interest due to the creation of sources of short high-power light pulses, the lasers. As the laser radiation passes through condensed media, additional refraction occurs in the latter, due to heating of the substance by absorption of radiant energy. This phenomenon may be utilized as a research tool. Thus in[2] a method was proposed and developed for the measurement of small local absorption coefficients, which is based on high-speed interferometric recording of change of the refractive index. On the other hand, in most cases the same phenomenon leads to undesirable nonlinear effects such as self-focusing or defocusing.[3] In either case the change of the refractive index is determined according to formula (1) with account taken of (2) or, for a sufficiently short time of action, according to the same formula (1) with account taken of the time dependence of ρ (nonstationary process). It is obvious that for a correct solution of problems of the type described above knowledge of each of the terms in the expression under the integral sign in (1) is indispensable.

Attempts at indirect determination of the quantity $(\partial n/\partial T)_\rho$ from measured parameters are known in the literature. Thus in[4] $(\partial n/\partial T)_\rho$ is calculated from the parameter $(\partial n/\partial P)_S$ measured in that work and from the parameters $(\partial n/\partial T)_P$, $(\partial T/\partial P)_S$, and β_S taken from the literature (P – pressure, S – entropy, β_S – isentropic compressibility). This method requires the knowledge of several isentropic constants, the measurement of which represents an independent, experimentally rather complex task. Besides, this method, applied in[4] to liquids, is hardly suitable for solids. In another paper[5] a method of calculating $(\partial n/\partial T)_\rho$ from the experimental ratio of intensities of the Raman and Rayleigh scattering is proposed. But since the square of the dipole polarizability appears in the expression for the intensity of the Raman scattering, $(\partial n/\partial T)_\rho$ can be computed only without a determination of its sign.

EXPERIMENTAL TECHNIQUE OF DIRECT MEASUREMENT OF $(\partial n/\partial T)_\rho$ AND ITS SUBSTANTIATION

Since the change produced in the refractive index by thermal expansion is established at the speed of sound in the substance under study, it follows from formula (1) that for the recording of the purely thermal effect the total time of heating and measuring Δn must be chosen to satisfy the condition

$$\left|\alpha\rho\left(\frac{\partial n}{\partial\rho}\right)_T\right| \ll \left|\left(\frac{\partial n}{\partial T}\right)_\rho\right| . \qquad (3)$$

The diagram of an experimental setup which allows to record the thermal variation of the refractive index is presented in Fig. 1. In this arrangement the substance is heated by the radiation of a neodymium laser with the following characteristics: spectrum–one longitudinal mode, pulse width 3×10^{-9} sec, maximum energy 0.3 J. With the help of the lens L (focal distance 150 cm) the radiation was focused into the cell C with the substance studied. The length of the cell was 7 cm, the effective radius of the laser beam at entry into the cell 0.2 cm, and the energy distribution close to the Gaussian. The variation of the refractive index was recorded with the Michelson interferometer M. For the illumination of the interferometer a part of the radiation of the neodymium laser was used, transformed into the second harmonic ($\lambda = 5300$ Å) with the help of the crystal KDP. The illumination pulse was delayed 3×10^{-9} sec relative to the heating pulse in order to exclude the influence of the electronic Kerr effect.

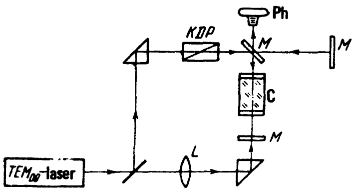

FIG. 1. TEM_{00}-neodymium laser, KDP-crystal transforming the radiation of $\lambda = 10600$ Å into radiation of $\lambda = 5300$ Å, L-lens focusing the neodymium laser radiation inside the investigated substance, M-mirrors of the Michelson interferometer, C-cell with the substance studied, Ph-camera.

The interference pattern was photographed by means of the camera Ph with its shutter open.

The basic working formula is of the simple form

$$\left(\frac{\partial n}{\partial T}\right)_\rho = \frac{C_v\lambda m}{2W\varkappa}, \qquad (4)$$

where C_v is the heat capacity at constant volume, λ the wavelength of the illuminating radiation, m the relative shift of the interference fringe at the maximum, W the energy density in the peak, and $\varkappa$ dimensionless constant that depends on the absorption coefficient of the substance and on the geometry of the laser beam inside the cell. The formula (4) allows to estimate the sensitivity of the method and the measurement error. These estimates yield: $(\partial n/\partial T)_\rho^{\min} \approx 0.25\times10^{-5}$ with a measurement error (±0.20%). To estimate the error of the method it is necessary to write the condition (3) with allowance for the specific geometry of the laser beam. Thus, for a Gaussian energy distribution the error of the method is

$$\Delta\left(\frac{\partial n}{\partial T}\right)_\rho \leqslant \alpha\rho\left(\frac{\partial n}{\partial\rho}\right)_T \frac{\Delta r r_0}{R^2}, \qquad (5)$$

where $r_0 = v\tau$ (v is the speed of sound, and τ the total time of measurement), Δr, is the accuracy with which the maximum of the interference pattern is determined. R is the effective radius of the laser beam at entry into the cell. Calculations using the formula (5) showed that the error of the method does not exceed (–10%).

The formula (4) contains C_v, the heat capacity at constant volume. In those cases when $(C_p - C_v) \sim C_p$, where C_p is the heat capacity at constant pressure, C_v was found from the well-known thermodynamic relation

$$C_v = C_p - \frac{\alpha^2 T}{\beta_T},$$

where β_T is the isothermal compressibility.

In working out the technique water was chosen as the standard substance. Water is the only substance for which $(\partial n/\partial T)_\rho$ can be measured in a static experiment. At temperatures near 4°C the co-efficient of thermal expansion of water becomes zero and, consequently, $dn/dT = (\partial n/\partial T)_\rho$ at this temperature. Figure 2 shows plots of the temperature dependence $\alpha(T)$ and $dn/dT(T)$ for water.[6]

RESULTS OF THE EXPERIMENT AND THEIR DISCUSSION

A typical interference pattern for water is given in Fig. 3 (top). The same figure (bottom) shows the distribution of energy density in the laser beam as it enters the cell. The closed curves are lines of equal energy density in relative units. The quantity $(\partial n/\partial T)_\rho$ for water was measured in a wide temperature interval (2 – 95)°C (see Fig. 2). It was established that in this temperature range $(\partial n/\partial T)_\rho$ for water does not depend on temperature and equals (-1.93×10^{-5}) deg^{-1}. An investigation of the influence of various admixtures in water on the quantity $(\partial n/\partial T)_\rho$ was likewise conducted.

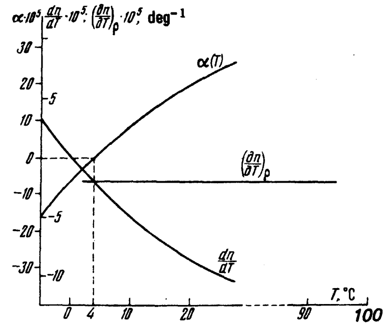

FIG. 2. Graphs of temperature dependence of the coefficient of thermal expansion α, the total derivative dn/dT [6] and the partial derivative $(\partial n/\partial T)_\rho$.

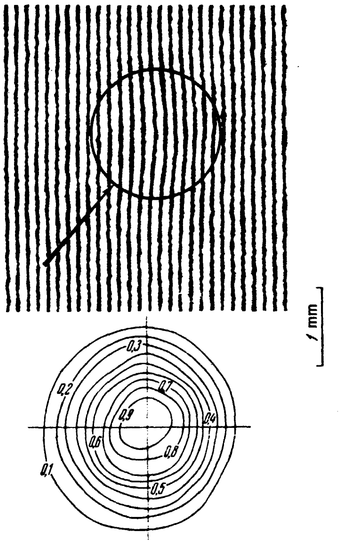

FIG. 3. A typical interference pattern for water (top) and distribution of energy density in the laser beam at entry into the cell (bottom) reverses sign, and attains the value $\approx 4\times 10^{-3}$deg^{-1} for concentrations near to the concentration of the saturated solution.

The salts $CuSO_4$ and $NiSO_4$ were used as strongly absorbing admixtures. It was found that when the concentration of these admixtures is increased to 10^{-2} g/cm^3 and the absorption coefficient grows correspondingly from 0.14 cm^{-1} to 0.5 cm^{-1}, the quantity $(\partial n/\partial T)_\rho$ remains constant. At the same time the sensitivity of the method increased (at a given cell length). The salt NaCl was used as a weakly absorbing admixture. The results of measurements of $(\partial n/\partial T)_\rho$ as a function of the NaCl concentration are shown in Fig. 4. It is seen that beginning with the concentration $\approx 10^{-2}$ g/cm^3 the quantity $(\partial n/\partial T)_\rho$ decreases in absolute value, passes through zero at the concentration 6×10^{-2} g/cm^3.

By analogy with water, we carried out an investigation of the influence on $(\partial n/\partial T)_\rho$ of strongly absorbing admixtures in optical glass. Glasses of the brand SZS were chosen with various concentrations of admixtures. The absorption coefficient of these glasses varied within the limits 2 cm^{-1} – 18.6 cm^{-1}. It was found that $(\partial n/\partial T)_\rho$ does not depend on the concentration of admixtures (within the limits indicated above). Since the basis of SZS glasses is the optical glass K-8, it is apparently possible to obtain the value of $(\partial n/\partial T)_\rho$ for that glass by linear extrapolation of the data for SZS glasses.

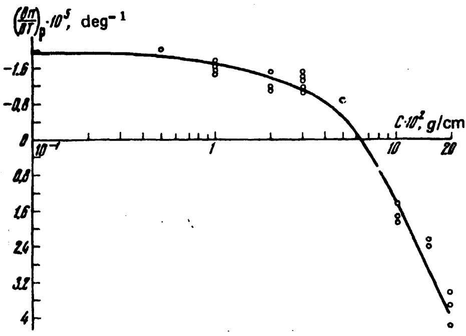

FIG. 4. Graph of the dependences of $(\partial n/\partial T)_\rho$ of an aqueous solution of NaCl on the NaCl concentration.

TABLE I.

Substance	$\alpha\cdot 10^5$	$\frac{dn}{dT}\cdot 10^5$	$(\partial n/\partial T)_\rho\cdot 10^5$	
			calculation	experiment
Water	23.8	−9.65	−1.97	−1.93
Chloroform	127.7	−58.8	+0.74	+0.90
Ethyl ether	165.1	−58.6	−0.78	−0.42
Glycerine	51.0	–	–	−0.60
K-8	2.2	+0.31	–	+0.92

In the same way measurements of $(\partial n/\partial T)_\rho$ for some other substances were carried out. Results of these measurements are presented in Table I. In the same table results of calculations based on the data of paper[4] are given. It is seen that, for instance in the case of ethyl ether, a discrepancy by a factor of almost two was observed. This discrepancy is evidently connected with the circumstance that, as is seen from formula (1), the quantity $(\partial n/\partial T)_\rho$ is obtained as a difference of two quantities and in the case of ethyl ether these are sufficiently close to each other in absolute value. The accuracy with which $(\partial n/\partial T)_\rho$ was measured in work[4] turns out to be insufficient.

Summarizing the results of the present work we may draw the following conclusions. A technique for direct measurement of the optical constant $(\partial n/\partial T)_\rho$ has been proposed and developed for the first time. Investigations of the dependence of $(\partial n/\partial T)_\rho$ on the temperature and on the concentration of various admixtures have been carried out for water. It has been shown that in the temperature range (2 – 95)°C the quantity $(\partial n/\partial T)_\rho$ does not depend on temperature. Nor does addition of absorbing admixtures in concentrations up to 10^{-2} g/cm^3 influence the quantity $(\partial n/\partial T)_\rho$. It has been established that in an aqueous solution of NaCl the quantity $(\partial n/\partial T)_\rho$ varies both in absolute value and in sign within the limits $(-1.9 - +4)\times 10^{-5}$ deg^{-1}. The values of $(\partial n/\partial T)_\rho$ were measured for a number of other substances.

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