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Angular Dependence of Rayleigh Light Scattering, Measured with a He-Ne Gas Laser

The angular dependence of Rayleigh light scattering is measured using a 15 mW He-Ne gas laser, presenting the advantage of an incident strongly monochromatic, collimated, plane-polarized beam of a relatively high spectral power density as compared with classical light sources.

The depolarization ratios and intensity components of light scattered throughout the angular distribution ranging from 50° to 130° have been measured for vertical and horizontal polarization of the electric vector of incident light in the following liquids: benzene, toluene, o-, m-, and p-xylene, cyklohexane, carbon tetrachloride, and chloroform.

Rayleigh coefficients of isotropic and anisotropic scattering, squared effective anisotropies of molecular optical polarizabilities, and angular correlation parameters have been calculated for these liquids with respect to the known absolute value of the Rayleigh coefficient for benzene at $\lambda = 632.8$ nm.

1. Expressions for the Rayleigh coefficient and depolarization ratio

The light scattering ability of medium irrespective of the experimental conditions is defined by its Rayleigh coefficient:

$$R_c = \frac{I_c R_0^2}{I_0 V}, \qquad (1)$$

where I_c is the total light intensity scattered in a given direction, R_0 the distance from the (small) scattering volume V to the observation point, and I_0 the intensity of the incident light beam.

The formula for the Rayleigh coefficient given by KIELICH in [1] is:

$$R_c(90) = \left(\frac{\pi^2}{10\lambda^4}\right)(5F_{is}+13F_{anis}), \qquad (2)$$

which may be generalized for all states of light polarization and all scattering angle to the form:

$$_iR_c^s(\Theta) = \frac{\pi^2}{5\lambda^4}\,[5F_{is}\cos_i^2\Omega_s + \\ + (3+\cos_i^2\Omega_s)F_{anis}], \qquad (3)$$

$_i\Omega_s$ is the angle between the electric field vectors of the incident wave (E_i) and scattered wave

(E_s). F_{is} and F_{anis} are commonly referred to as the molecular factors of isotropic and anisotropic light scattering, respectively, and can be calculated theoretically by statistical mechanics, and determined experimentally from the absolute values of the Rayleigh coefficient [2, 3]:

$$F_{is} = kT\beta_T\,N\left(\frac{\partial\varepsilon}{\partial N}\right)_T^2, \qquad (4)$$

$$F_{anis} = \left(\frac{\delta^2 G}{N}\right)\left(N\frac{\partial\varepsilon}{\partial N}\right)_T^2, \qquad (5)$$

where k -- Boltzmann's constant; T — absolute temperature; ε — electric permittivity of the medium; N — number of molecules per unit volume; δ — anisotropy in optical polarizability of the isolated molecule; β_T — isothermal compressibility coefficient of the medium; and $G = 1 + J_k$ — the parameter of angular correlations of the molecules [2].

The most usual procedure consists in measurements with the incident light wave polarized either vertically, or horizontally, to the plane of observation. For plane-polarized incident light, and plane-polarized components of scattered light, four Rayleigh coefficients V_v, H_v, V_h, H_h, can be measured. The notation used being due to Krishman, where the capitals H and V stand for horizontal and vertical polarization states of scattered light component,

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while the subvertices h and v represent the respective states of the incident light. By virtue of Eq. (3), the following relations can be derived easily for polarized incident light [1, 3]:

$$V_v(\Theta) = \frac{\pi^2}{5\lambda^4}(5F_{is}+4F_{anis}),_i \Omega_s = 0^\circ, \quad (6)$$

$$V_h(\Theta) = H_v(\Theta) = \frac{3\pi^2}{5\lambda^4}F_{anis} \,_i \Omega_s = 90^\circ, \quad (7)$$

$$H_h(\Theta) = \frac{\pi^2}{5\pi^4}[5F_{is}\cos^2\Theta + (3+\cos^2\Theta)F_{anis}], \quad (8)$$

whereas for non-polarized incident light (subscript n) we have:

$$V_n(\Theta) = V_v(\Theta) + V_h(\Theta) = \frac{\pi^2}{5\lambda^4}(5F_{is}+7F_{anis}), \quad (9)$$

$$H_n(\Theta) = H_h(\Theta) + H_v(\Theta)$$
$$= \frac{\pi^2}{5\lambda^4}[5F_{is}\cos^2\Theta + (6+\cos^2\Theta)F_{anis}], \quad (10)$$

and the total Rayleigh coefficient for natural incident light is:

$$R_n(\Theta) = [V_v(\Theta)+V_h(\Theta)+H_v(\Theta)+H_h(\Theta)]/2$$
$$= \frac{\pi^2}{10\lambda^4}[5(1+\cos^2\Theta)F_{is} + (13+\cos^2\Theta)F_{anis}]. \quad (11)$$

From Eqs. (6)–(11) it follows that the Rayleigh coefficient components H_h and H_n are the functions of the scattering angle, whereas the others are angle independent constants.

Eq. (11) can be re-written as:

$$R_n = R_{is}+R_{anis}, \quad (12)$$

where:

$$R_{is} = \frac{\pi^2}{2\lambda^4}(1+\cos^2\Theta)F_{is}, \quad (13)$$

and

$$R_{anis} = \frac{\pi^2}{10\lambda^4}(13+\cos^2\Theta)F_{anis}. \quad (14)$$

In the experiments on light scattering, besides the Rayleigh coefficients, quantities referred to as depolarization ratios of scattered light have been determined. For polarized incident light, the depolarization ratios are given by the Eqs. [1, 3]:

$$= D_v(\Theta) = \frac{H_v(\Theta)}{V_v(\Theta)} = \frac{3F_{anis}}{5F_{is}+4F_{anis}}, \quad (15)$$

$$D_h(\Theta) = \frac{V_h(\Theta)}{H_h(\Theta)}$$
$$= \frac{3F_{anis}}{5F_{is}\cos^2\Theta+(3+\cos^2\Theta)F_{anis}}, \quad (16)$$

and for unpolarized incident light by

$$D_n(\Theta) = \frac{H_n(\Theta)}{V_n(\Theta)} = \frac{2D_v}{1+D_v}$$
$$= \frac{5F_{is}\cos^2\Theta+(6+\cos^2\Theta)F_{anis}}{5F_{is}+7F_{anis}}, \quad (17)$$

when calculating R_{is} and R_{anis}, it is convenient to refer to the experimentally measured depolarization ratio D_n. For $Q = 90^\circ$, we have:

$$R_{is}(90) = R_n(90)\frac{6-7D_n(90)}{6+6D_n(90)}, \quad (18)$$

where

$$\frac{6+6D_n(90)}{6-7D_n(90)}$$

is the Cabannes coefficient.

The quantities determined from Eq. (18) can be compared with the theoretical values calculated from the [5]:

$$R_{is} = \frac{\pi^2}{2\lambda^4}KT_{\beta_T}\left(N\frac{\partial\varepsilon}{\partial N}\right)_T^2, \quad (19)$$

R_{anis} is given by the following relations:

$$R_{anis}(90) = R_n(90)\frac{13D_n(90)}{6+6D_n(90)} \quad (20)$$

and

$$R_{anis}(90) = \frac{13\pi^2}{10\lambda^4}\frac{\left(N\frac{\partial\varepsilon}{\partial N}\right)^2}{N}\delta^2(1+J_k), \quad (21)$$

where δ is the optical anisotropy of the isolated molecule, the squared value of the latter being given by:

$$\delta^2 = \frac{(a_1-a_2)^2+(a_2-a_3)^2+(a_3-a_1)^2}{2(a_1+a_2+a_3)^2}. \quad (22)$$

Here

a_1, a_2, a_3 are the polarizabilities in the directions of the principal axes of the molecule,

J_k — is the parameter of angular correlations, for axially symmetric molecules given by the expression [13]

$$J_k = \frac{\varrho}{2V} \int\int (3\cos^2\theta_{pq}^{-1}) g(\tau_p, \tau_q) d\tau_p d\tau_q, \quad (22a)$$

where:

$g(\tau_p\tau_q)$ — is the binary correlation functions,
θ_{pq} — the angle between the axes symmetry of two interacting molecules p and q having the configurations τ_p and τ_q.

The angular correlation parametr J_k can be determined numerically by a direct measurement of the H_v component of anisotropic light scattering [6]. By (7),

$$H_v = \frac{3\pi^2}{5\lambda x^4} F_{anis}.$$

The anisotropic scattering factor F_{anis} can be expressed in the form [1], [3]:

$$F_{anis} = \frac{\delta^2(1+J_k)}{N} \left(N\frac{\partial\varepsilon}{\partial N}\right)^2. \quad (23)$$

The procedures leading to $\left(N\frac{\partial\varepsilon}{\partial N}\right)$ have been discussed by DEŽELIĆ [3], COUMOU [5], SHAKHAPORONOV [7], and KIELICH and PIECZYŃSKA [8] for various models. The values closest to the experimental ones are obtained from the expression [3]:

$$N\frac{\partial\varepsilon}{\partial N} = 4\pi Na', \left[\frac{(n^2+2)}{3}\right], \quad (24)$$

where a' is the effective polarizability of the molecules in the medium, and n its refractive index of the latter. The optical anisotropy δ^2 can be expressed in terms of the effective anisotropy as follows:

$$\delta^2 = \gamma'^2/9a'^2 \quad (25)$$

with γ'^2 defined by the relation:

$$\gamma'^2 = \frac{(a_1-a_2)^2+(a_2-a_3)^2+(a_3-a_1)^2}{2}. \quad (26)$$

Thus Eq. (23) may be presented in the following form:

$$F_{anis} = \frac{16\pi^2}{9} N\gamma'^2(1+J_k)\left(\frac{n^2+2}{3}\right)^2, \quad (27)$$

and the H_v component in the form

$$H_v = \frac{16\pi^2}{15\lambda^4} N\gamma'^2(1+J_k)\left(\frac{n^2+2}{3}\right)^2. \quad (28)$$

From the Eqs. (14), (27) and (28) the anisotropic part of the Rayleigh coefficient may be simply presented as:

$$R_{anis} = \frac{13H_v}{6}. \quad (29)$$

Denoting

$$\gamma'^2(1+J_k) = \Gamma^2 \quad (30)$$

Eq. (28) takes the form

$$H_v = \frac{16\pi^2}{15\lambda^4}\left(\frac{n^2+2}{3}\right)^2 N\Gamma^2, \quad (31)$$

Γ^2 is referred to as the effective anisotropy of short-range optical polarizability.

Once the scattered intensity component H_v is measured, and the refractive index of the medium at $\lambda = 632.8$ nm and the number of molecules per unit volume are available, the squared effective anisotropy of optical polarizability of the molecules is obtained from the relation (31):

$$\Gamma^2 = \frac{H_v}{\frac{16\pi^2}{15\lambda^4}\left(\frac{n^2-2}{3}\right)^2 N}. \quad (32)$$

When the squared effective anisotropy of optical polarizability of the molecules Γ^2 is known and the anisotropy of linear polarizability of the isolated molecule γ'^2 is available from the literature, Eq. (30) permits to calculate the value of the angular correlation parameter J_k.

2. Method of measurements

The angular dependence of light scattering was measured with the device shown in Fig. 1. The light source was a He-Ne laser, operating in a fundamental mode and generating a beam of light wavelength $\lambda = 632.8$ nm and power 15 mW. The laser consisted of a capillary tube of internal diameter 1.5 mm and length 1.5 m with windows at Brewster angle, and two dielectric mirrors forming a plane-concave optical resonator. The plane output mirror had a transmittivity of 2.6% for $\lambda = 632.8$ nm, whereas the concave mirror of curvature radius
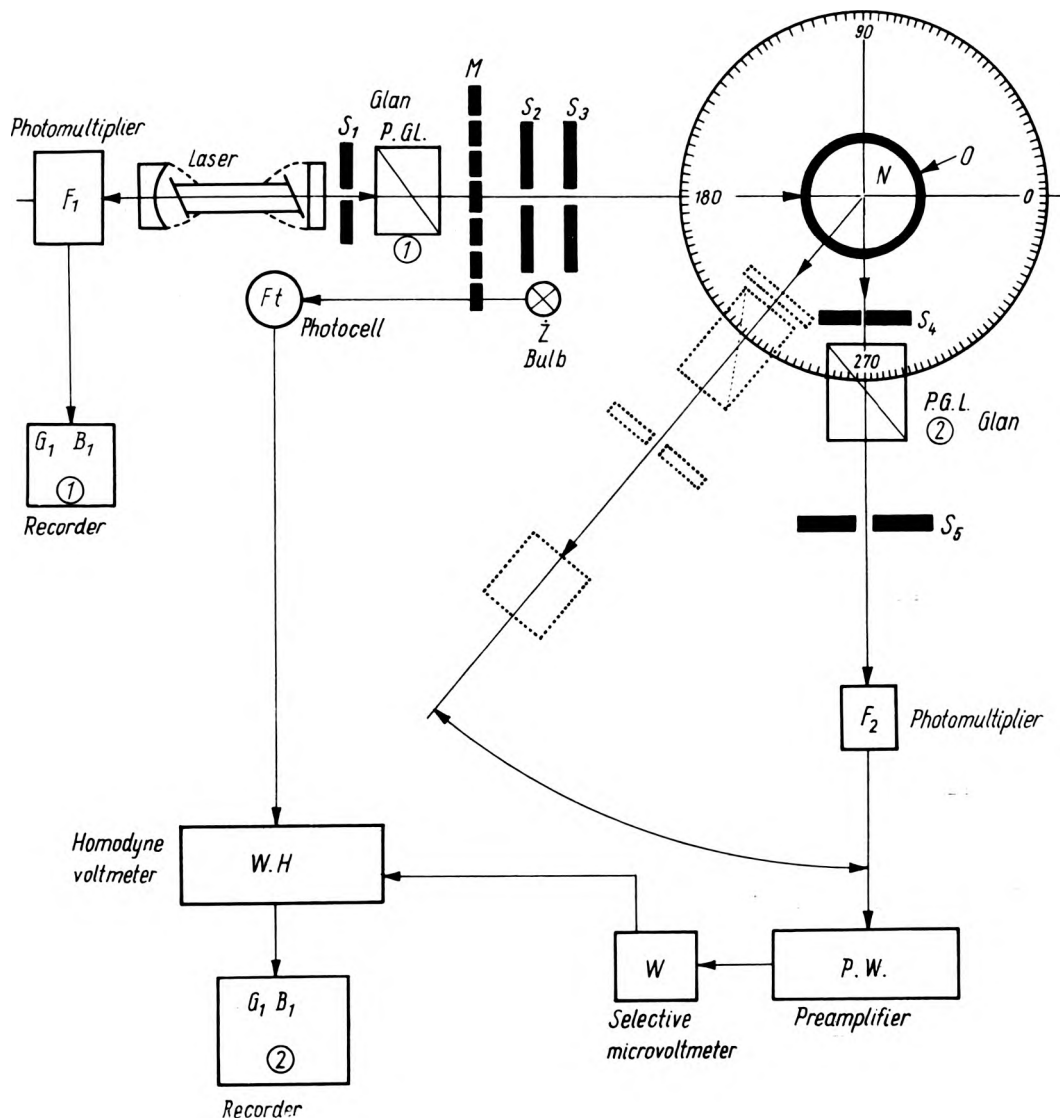
Fig. 1. Arrangement used for the measurement of the angular dependence of scattered light intensity

3.5 m transmitted 0 %. With this setup the divergence of the beam did not exceed $0.5 \cdot 10^{-4}$ rad. The laser tube was connected by a concentric cable to a DC current supply. To improve the stabilization of laser power stabilization, an RC filter was included in the supply circuit ($R = 240$ kΩ, $C = 360$ μF). The supplied voltage amounted to 8 kV at maximal current 10 mA. The laser beam power was measured on the side of the 0 % transmission mirror by means of an FEU-22 photomultiplier (on preattenuation of the light flux) and recorded graphically.

The laser tube was disposed so that the electric vector of the beam vibrated at an angle of 45° to the vertical. Rotation of the Glan polarization prism permitted to set the electric vector of the incident beam at 0° or, 90° with respect to the plane of observation. With crossed polarization prisms in the paths

of the incident and detected beams, extinction in the detection circuit was complete, and no signal was recorded by the measuring system. Rotation of the prisms was operated with an accuracy of l'.

The sample cuvette consisted of a Pyrex glass tube with a wall thickness of 1 mm and internal diameter of 46 mm closed off with a ground glass plug. In order to eliminate parasitic stresses, the glass was kept in a furnace at 420°C for 90 min and tested optically with a polarimeter.

Parasitical scattering by the walls of the cuvette was negligible owing to the small diameter of the incident beam (1 mm), the relatively larger diameter of the cuvette (46 mm) and careful cleaning of its walls. In the absence of the liquids, no signal was recorded by the measuring system. The cuvette was enclosed
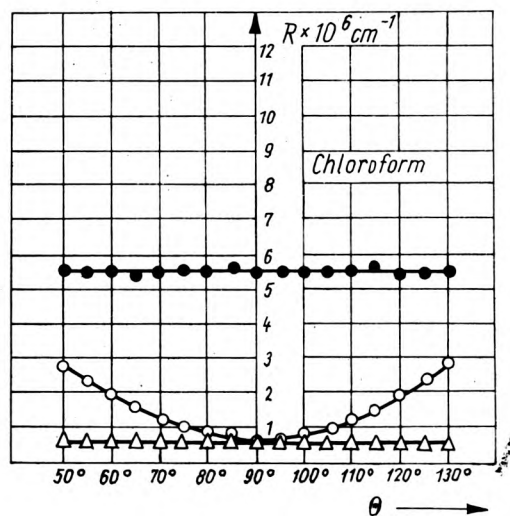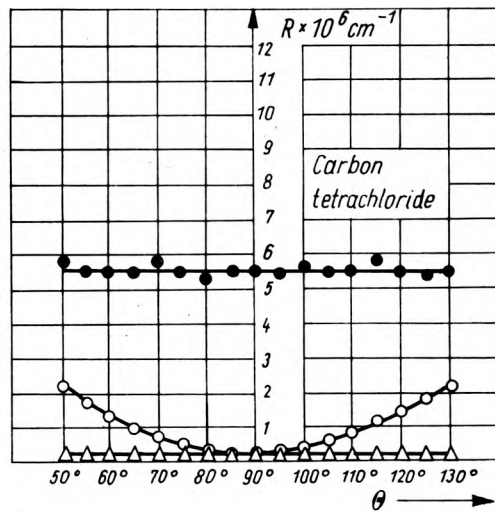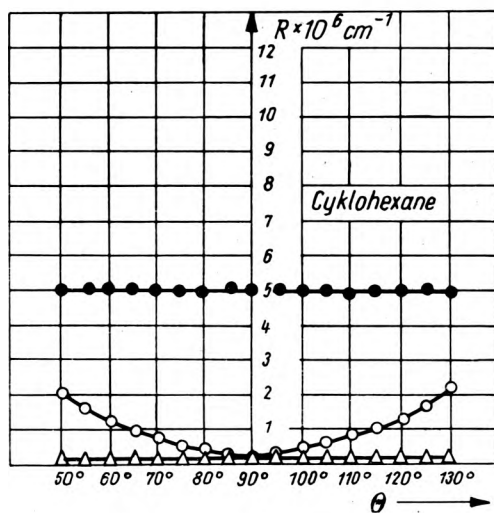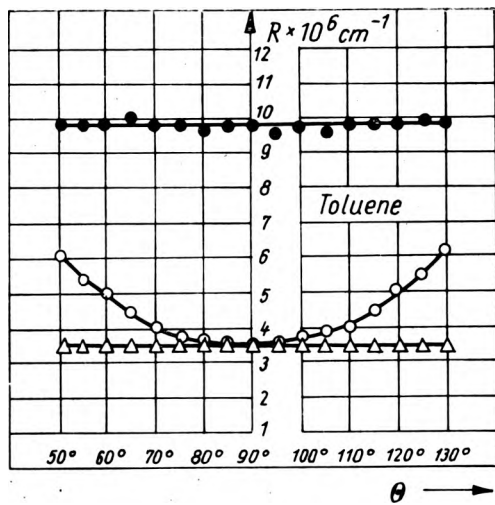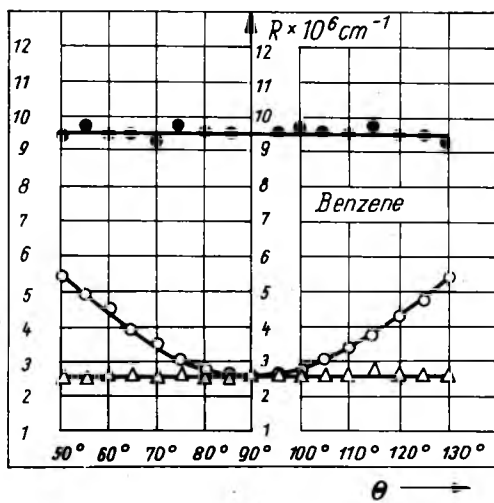
Figs. 2-6. Angular dependence of Rayleigh coefficient. ● — V_v, ○ — H_h, △ — $H_v = V_h$
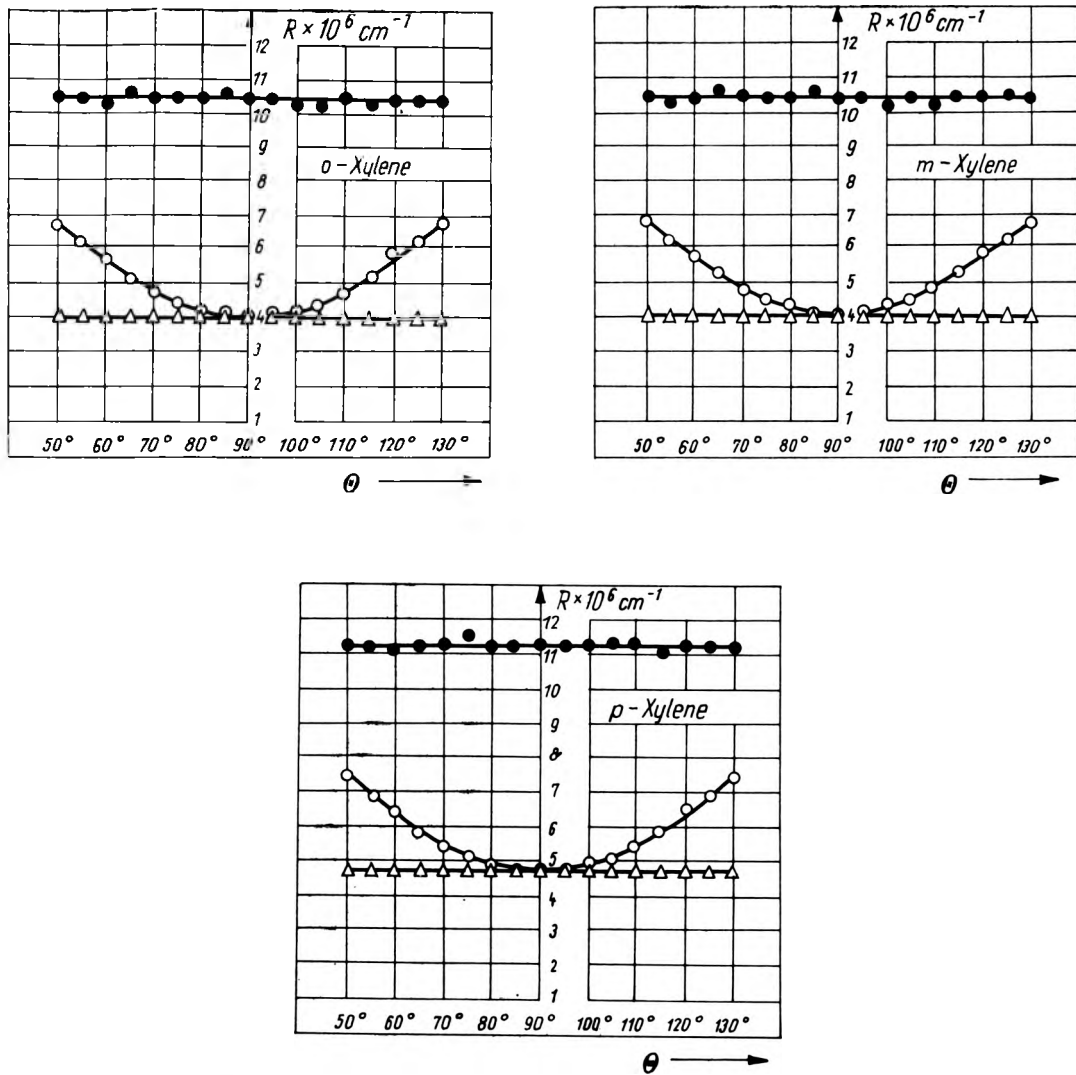
Figs. 7-9. Angular dependence of Rayleigh coefficient. ● — V_v, ○ — H_h, △ — $H_h = V_h$

in a light-tight metal shield blackened with lustreless paint.

Because of the waekness of the scattered light signals and the necessity of its amplification, the incident beam was modulated mechanically with the frequency 10 Hz by means of a six-aperture chopper rotated by an SS-1 synchronous motor at 60 rev/min. The detecting system was mounted on a rotatable arm, which can be put into motion on a metal support of 200 mm diameter by means of a toothed wheel. The observation direction of scattered light with respect to that of the incident beam was established with an accuracy of 30'. At a 35 mm distance from the centre of the scattering volume, a rectangular slit (S_4) 2 mm wide, closely adhering to the metal shield of the cuvette, was disposed. The scattered light traversed a Glan polarization prism (P. Gl.$_{(2)}$),

rotatable with an accuracy of l', and a 2 mm wide slit (S_5) distant by 75 mm from the centre of the system. The scattered light signal was recorded with an FEU-17 A photomultiplier distant by 150 mm from the centre of the scattering volume.

The signal from the photomultiplier was transferred by a 203-50 type preamplifier to a 203 type selective microvoltmeter (W) with 10 Hz appliance and a 202 B type homodyne voltmeter (W-H-), both made by "Unipan" (Warsaw). As a reference signal for the homodyne voltmeter we have used that from a photocell (Ft) illuminated by the light flux of a 6 V bulb (Z) placed in the path of the mechanical modulator of the incident beam. The signal from the homodyne voltmeter was fed to a DC current recording device $G_1 B_{1(2)}$.

3. Results of measurements and conclusions

The liquids used in our measurements were either of *pro analysi* grade, and subjected to triple distillation, or spectrally pure, distilled directly into the measuring cuvette through a G-4 millipore filter. Measurements were performed for the following liquids:

Benzene — spectrally pure, from the British makers B. D. H.,

Toluene — spectrally pure, from Merck,

Cyklohexane — spectrally pure, Polish made,

Carbon tetrachloride — of *pro analysi* grade, produced in Poland,

Chloroform — of *pro analysi* grade, produced in Poland,

Ortho-, meta-, and para-xylene — of *pro analysi* grade, from the British firm B. D. H.

Chemical purity of the liquids was assessed by refractive index measurements for the yellow line of sodium by an Abbe' refractometer with the accuracy to appr. four decimals. The results were compared with the tabulated data given in [9]. The required mechanical purity was ensured by a direct distillation into the cuvettes.

The measurements of scattered radiation intensity, performed in this work bear a relative character. Therefore, only the corrections relevant in this respect have been taken into account, i.e. for solid angle (related to refractive index of the liquid), for variations in scattering volume (while studying the angular dependence of scattering), and for differences in the photomultiplier sensitivity to polarization direction of the recorded beam [10, 11]. The corrections defined by geometrical parameters of the device remained for all liquids the same, thus being irrelevant in the relative measurements. By means of the arrangement described above, the angular dependences of Rayleigh coefficients have been determined for the organic liquids like benzene, toluene, cyclohexane carbon tetrachloride, chloroform, as well as for o-, m-, and p-xylene, and plotted in Figs 1–8. The values of the Rayleigh coefficients are given in units of $[cm^{-1}]$. We referred to the total Rayleigh coefficient for benzene, given in Ref. 12 for the wavelength $\lambda = 632.8$ nm:

$$R_n = 8.765 \cdot 10^{-6} \ [cm^{-1}].$$

Table includes the values of R_{is}, R_{anis}, D_v, D_n, Γ^2 and J_k calculated from our measurements of the scattered light components V_v and H_v, and the comparative values taken from the available literature.

On the graphs plotted for the angular dependence of scattering, the measurement points are denoted by little circles Figs. 2–9, whereas full line represents the theoretical curve, calculated from the following relation [11]:

$$H_h = H_v + (V_v - H_v)\cos^2 \Theta, \qquad (33)$$

The values of the Rayleigh coefficient and depolarization ratio obtained experimentally as compared with literature data of total Rayleigh coefficients and depolarization ratios

	$H_v \cdot 10^6$ cm^{-1} ex-per.	$V_v \cdot 10^6$ cm^{-1} exper.	$H_h \cdot 10^6$ cm^{-1} ex-per.	$V_h \cdot 10^6$ cm^{-1} ex-per.	$R_{is} \cdot 10^6$ cm^{-1}	$R_{anis} \cdot 10^6$ cm^{-1}	$R_n \cdot 10^6$ cm^{-1} exper.	R_n Literature	D_v ex-per.	D_n	D_n Literature
Benzene	2.662	9.545	2.662	2.662	2.998	5.767		8.765 [12] 8.35 [14] 8.64 [15]	0.279	0.436	0.435 [14] 0.423 [15]
Toluene	3.543	9.912	3.543	3.543	2.644	7.801	10.445	10.31 [12] 8.77 [14]	0.357	0.526	0.528 [14]
Cyclohexane	0.138	4.956	0.138	0.138	2.387	0.297	2.684	2.54 [15]	0.028	0.054	0.059 [14] 0.047 [15]
Carbon tetrachloride	0.125	5.507	0.125	0.125	2.671	0.270	2.941	3.1 [14] 3.065 [15]	0.023	0.044	0.045 [16] 0.038 [14] 0.042 [15]
Chloroform	0.652	5.580	0.652	0.652	2.356	1.412	3.768	3.87 [15]	0.116	0.208	0.204 [14] 0.215 [15]
ortho-xylene	4.038	10.463	4.038	4.038	2.538	8.751	11.289		0.386	0.557	
meta-xylene	4.093	10.463	4.093	4.093	2.527	8.844	11.371		0.389	0.560	
para-xylene	4.736	11.197	4.736	4.736	2.435	10.267	12.702		0.423	0.595	

The good agreement between the experimental and theoretical curves shows that angular dependence of the Rayleigh coefficient in the liquids investigated is fulfilled.

Since the molecules are much smaller than $\lambda/20$, the angular distribution of scattered light components is symmetric about the angle $\Theta = 90°$.

The values D_n, R_n, Γ^2 and J_k calculated from our measurements and presented in Table are comparable with the literature data. Angular correlations for these liquids obtained experimentally are close to the values calculated theoretically from the relation [13]:

$$G = 1 + J_k = \frac{10 D_n R_{\text{is}}}{a^2 \delta^2 (6 - 7 D_n)} \left(\frac{\lambda}{2\pi}\right)^4 \left(\frac{3}{n^2 + 2}\right)^2,$$

but highly differ from those obtained from the formula [13]:

$$1 + J_k = \frac{5 N T K \beta_T D_n}{\delta^2 (6 - 7 D_n)}.$$

An important property of the parameter J_k is that it is positive or negative, according to the type of molecular orientation. Thus, if $J_k < 0$, there is a tendency to a perpendicular orientation of the molecules: if $J_k > 0$, the orientation tends to be parallel; while $J_k = 0$ indicate that molecular correlation does not occur.

From the performed calculations of the angular correlation parameter, based on measurements of light scattering, it follows that the molecules of benzene, toluene and cyclohexane are found to exhibit a tendency mutually perpendicular orientation, while those of chloroform and o-, m-, and p-xylene tend to orientate parallely. The molecules of carbon tetrachloride appear to be angularly uncorrelated.

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La mesure d'une dépendance angulaire de la dispersion de lumière de Raleigh en utilisant le laser gazeux He-Ne

Dans cet ouvrage on a présenté des recherches menées à l'aide du laser gazeux He-Ne dont la puissance était de 15 mW. L'utilisation de ce laser a permis à une analyse de lumière dispersée pour un faisceau lumineux monochromatique, parallel, polarisé linéairement avec une densité de puissance spectrale relativement haute en comparison avec des sources de lumière classiques.

On a mesuré les niveaux de dépolarisation de lumière et les composantes de la lumière dispersée avec une distribution angulaire de 50° à 130°, pour un vecteur électrique parallèlement polarisé de l'onde électrique incidente pour les liquides suivant: benzène, toluène, o-, m-, p-xylène, cyclohexane, tétrachlorure de carbone et chloroforme.

En se servant de la valeur absolue du facteur de Raleigh obtenu pour le benzène pour une longeur d'onde de 632,8 nm, on a calculé des facteurs de Raleigh de la dispersion isotrope et anisotrope, les carrées d'anisotropie efficace de la polarisation optique des molécules, et des paramèters de correlation angulaire de ces liquides.

Измерение угловой зависимости релеевского рассеяния света при употреблении газового лазера He-Ne

Описанные в настоящей работе исследования проводились при применении газового лазера He-Ne мощностью в 15 мвт. Применение лазера позволило исследовать рассеянный свет для падающего пучка, в значительной степени монохроматического, параллельного, линейно поляризованного, со сравнительно значительной плотностью мощности по сравнению с классическими источниками света.

Измерены степени деполяризации света и составляющие интенсивности рассеянного света по угловому распределению в диапазоне от 50 до 130°, для перпендикулярно и параллельно поляризованного электрического вектора падающей волны для следующих жидкостей: бензол, толуол, о-, т-, р-кислол, циклогексан, четыреххлористый углерод и хлороформ.

С использованием вычисленной абсолютной величиной коэффициента Релея для бензола при длине волны 632,8 нм рассчитаны коэффициенты Релея изотропного и анизотропного рассеяния, квадраты анизотропии оптической поляризуемости молекул и параметры угловых соотношений для этих жидкостей.

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