

which is inactive for the single-photon 0-i transition.

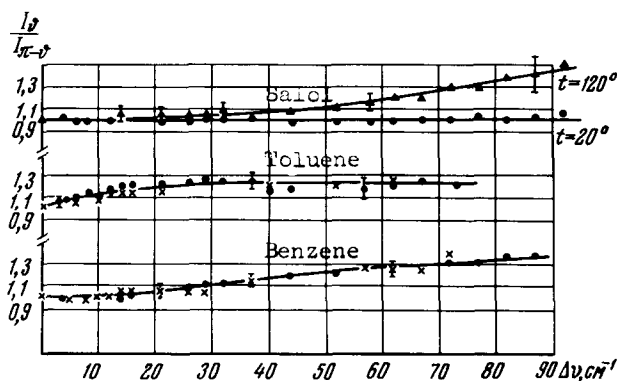
ANGULAR DISTRIBUTION OF INTENSITY IN THE THERMAL WING OF THE RAYLEIGH LINE IN LIQUIDS

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We have investigated the spectral distribution of the intensity of scattered light in the wing of the Rayleigh line [1,2] in the frequency interval 0 - 100 cm^{-1} from the undisplaced line in benzene, carbon disulfide, toluene, carbon tetrachloride, salol, and benzophenone at scattering angles 60, 90, and 120°.

The measurements were made with apparatus described earlier [2] by the following procedure: The fluorescence of a solution of quinine sulfate, whose indicatrix has spherical symmetry, was first registered at an angle 60°, followed by registration of the spectrum of the Rayleigh-line wing at the same angle and with other conditions constant, after which the fluorescence light was again registered. Similar operations were performed for 90 and 120°. After reduction of the spectrograms, the ratios $z_1(\omega) = (I_{60^\circ}(\omega)/I_{120^\circ}(\omega))$ and $z_2(\omega) = (I_{90^\circ}(\omega)/I_{120^\circ}(\omega))$ were calculated for each investigated liquid as functions of the frequency of the scattered light, and were multiplied respectively by the ratios $I_{\text{quinine}}(120^\circ)$ to $I_{\text{quinine}}(60^\circ)$ and $I_{\text{quinine}}(120^\circ)$ to $I_{\text{quinine}}(90^\circ)$ in order to exclude the possible difference in the scattering volumes of the investigated liquid at the indicated angles and the difference in the intensity of the exciting light. The error in the determination of the quantities $z_1(\omega)$ and $z_2(\omega)$, due to the difference between the refractive indices of the quinine sulfate solution and the investigated liquids, amounted to a fraction of one per cent and was disregarded. Benzene, toluene, and carbon disulfide were illuminated with x- and z-polarized light; the x-component was separated in the scattering light up to 40 cm^{-1} . The carbon tetrachloride, salol, and benzophenone were excited with natural light only.

The results of the experiment for some of the investigated liquids are shown in the figure, from which it is seen that for benzene the ratio $z_1(\omega)$ is equal to unity in the frequency interval from 0 to 20 cm^{-1} , and that from 20 cm^{-1} to 90 cm^{-1} the intensity of the light scattered forward exceeds the intensity of the back-scattered light. A similar relation for $z_1(\omega)$ is observed in the case of toluene at room temperature and salol at 120°C. For carbon disulfide, carbon tetrachloride, benzophenone and salol at



Frequency dependence of the ratio $z_1(\omega)$ in the wing of the Rayleigh line: • - exciting light z-polarized, × - exciting light x-polarized.

room temperature, a symmetrical indicatrix is observed.

The ratio $z_2(\omega)$ equals, within the limits of measurement error, 0.81 when the liquid is excited with polarized light and 0.98 for natural light; this is in good agreement with the deductions of the theory of light scattering as a result of that part of the anisotropy fluctuations which is described by a symmetrical anisotropy tensor [3]. Forward scattering becomes noticeable in benzene, toluene, and salol at $t = 120^\circ$ only at frequencies exceeding $\omega_{re} = 1/\tau_{re}$ (τ_{re} = time between two reorientations of the molecules [2,4]).

We can attempt to explain the results by assuming that a short-range orientation order and correlation of the oriented motion of the neighboring molecules at time intervals $t < \tau_{re}$, i.e., times during which the molecules excite rotational oscillations, exist in those liquids where a deviation of $z_1(\omega)$ from unity is observed. If this is so, then the light scattered by different molecules of the liquid is coherent and therefore, as a result of the interference, the forward scattering will be predominant only in the spectral region for which the rotational vibrations of the molecules are responsible ($\omega > \omega_{re}$).

In carbon disulfide and carbon tetrachloride there is apparently no short-range orientation order, and therefore $z_1(\omega) = 1$ for any scattered-light frequency.

The absence of asymmetry in the angular distribution of the intensity of the scattered light by viscous liquids at room temperature may mean from this point of view that the molecules have on the average a random orientation in the volume where the correlation in the oriented motion of the molecules is appreciable, although a short-range orientation order does exist in individual parts of this volume. The increase in the temperature of the viscous liquid decreases the regions in which correlation in the molecule motion exists, and perhaps there is already a short-range orientation order for these volumes. This possibly is why $z_1(\omega) > 1$ is obtained for salol at $t = 120^\circ$. Unfortunately, it is still impossible to describe quantitatively the ideas advanced here to explain the results.

In conclusion, the authors are sincerely grateful to I. L. Fabelinskii for direction of this work.

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