

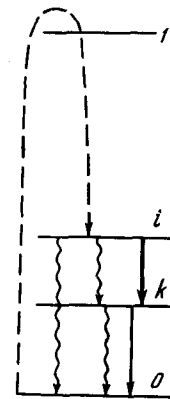
1) A. B. Mikhailovskiy has remarked that in the case of sufficient initial scatter in $V_{\parallel i}$ the stabilizing effect may appear in higher-order approximations in n_k , as a result of nonlinear Cerenkov radiation of the ions.

POSSIBILITY OF OBSERVING INDUCED INFRARED RADIATION IN RAMAN SCATTERING OF LIGHT

V. S. Gorelik, V. A. Zubov, M. M. Sushchinskii, and V. A. Chirkov
 P. N. Lebedev Physics Institute, USSR Academy of Sciences
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We discuss below a new mechanism of producing population inversion between vibrational or vibrational-rotational levels of molecules.

Let us consider a molecule with the level scheme indicated in the figure. We assume that: (i) the transition 0-i is allowed in the Raman spectrum and forbidden for the single-photon process; (ii) the transition 0-k, to the contrary, is forbidden for Raman scattering and allowed for the single-photon process; and (iii) the transition k-i is allowed for the single-photon process. These conditions are satisfied, for example, for vibrational transitions of all molecules possessing an inversion center. When conditions (i - iii) are satisfied, the level k will not become populated in the case of Raman scattering of light, so that the thermal distribution of the molecules over the vibrational levels may become disturbed and population inversion may occur. The gain per centimeter for the i-k transition near the generation threshold is



$$k = \frac{\lambda^2}{8\pi} \frac{A_{ik}}{\Delta\nu} g_i \left(\frac{N_i}{g_i} - \frac{N_k}{g_k} \right) = \frac{\lambda^2}{8\pi} \frac{A_{ik}}{\Delta\nu} g_{ik} \left(\frac{q_i}{w_i g_i} - \frac{N_k}{g_k} \right), \quad (1)$$

where g_i and g_k are the statistical weights of the upper and lower levels, λ is the wavelength of the i-k transition, N_i and N_k are the numbers of molecules per cm^3 at the upper (i) and lower (k) levels, A_{ik} is the Einstein coefficient for spontaneous emission, q_i is the rate of "pumping" of molecules to the upper level, w_i is the total probability of decay of this level, and $\Delta\nu$ is the line width of spontaneous emission in sec^{-1} . Far from resonance the absolute quantum yield of Raman scattering per molecule $\bar{\sigma}$ does not depend on the frequency of the exciting light, and then $q_i = \bar{\sigma} N_0 \bar{P} / \hbar \bar{\omega}_0$, where \bar{P} is the effective value of the power density of the incident radiation, $\bar{\omega}_0$ is the effective value of the circular frequency of the incident light, and N_0 is the number of molecules per cm^3 in the ground state. At sufficiently low temperatures the second term in (1) can be neglected, and then at a resonator length $l = 10$ cm and at a mirror reflection coefficient $R = 99\%$ we obtain the following necessary condition for generation:

$$k = \frac{\lambda^2 A_{ik} \bar{\sigma} N_0 \bar{P}}{8\pi \Delta\nu w_i \hbar \bar{\omega}_0} \geq 10^{-3}. \quad (2)$$

It is necessary to add to condition (2) the requirement of sufficiently rapid depletion of the lower working level, a condition satisfied in most cases [1].

For preliminary estimates we can assume for gases $\lambda = 10 \mu$, $N_0 = 3 \times 10^{19} \text{ cm}^{-3}$, $\bar{\sigma} = 2 \times 10^{-28} \text{ cm}^2$ [2], $A_{ik} = 10^2 \text{ sec}^{-1}$, $w_i = 10^2 \text{ sec}^{-1}$ [3] ¹⁾, $\Delta\nu = 3 \times 10^{10} \text{ sec}^{-1}$, $\hbar\omega_0 = 10^{-12} \text{ erg}$. We then obtain from (2) that generation is possible when $\bar{P} \geq 10^4 \text{ W/cm}^2$. For a liquid $N_0 = 3 \times 10^{22} \text{ cm}^{-3}$, $w_i = 10^8 \text{ sec}^{-1}$ [1] ²⁾ and from (2) it follows that generation necessitates $\bar{P} \geq 10^7 \text{ W/cm}^2$. Estimates made for a xenon lamp ($\tau = 10^{-3} \text{ sec}$, $\bar{P} \approx 10^5 \text{ W/cm}^2$) show that it is possible to obtain generation in the gas. The molecule density at the upper level turns out to be $N_i = 10^{13} \text{ cm}^{-3}$.

On approaching the electron-absorption bands, the absolute quantum yield of the Raman scattering of light increases by 4 - 5 orders [5,6]. The condition (2) is satisfied in this case when $\bar{P} \geq 0.1 \text{ W/cm}^2$ for gases and when $\bar{P} \geq 10^2 \text{ W/cm}^2$ for liquids. Assuming for the same light source $\bar{P} = 10^4 \text{ W/cm}^2$ in the resonance region, we obtain $N_i = 10^{17} \text{ cm}^{-3}$ in the case of gases and 10^{14} cm^{-3} for liquids, i.e., in both cases generation is possible at sufficiently low temperatures.

By using a ruby laser it is possible to obtain $\bar{P} = 10^7 \text{ W/cm}^2$ at a pulse duration $\tau = 10^{-8} \text{ sec}$. Usually for monochromatic radiation with such a power density one observes induced Raman scattering in liquids, for which the quantum yield can reach several times ten per cent, i.e., we can assume that $P = 0.1P_0$, where P is the total power of the induced Raman scattering and P_0 the power density of the exciting radiation. Then $N_i = 10^{16} \text{ cm}^{-3}$, which is much higher than the threshold value. This excitation mechanism is realizable in principle in crystals, too.

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¹⁾ For a gas we choose w_i to be the rate of decay of the vibrational level, since the short lifetime of the rotational levels leads only to a rapid establishment of the thermal equilibrium in the rotational-vibrational band. We note that in a gas population inversion can be produced between rotational sublevels of different vibrational levels even in the case when $N_i/N_k \leq 1$ [4], where N_i and N_k are the total numbers of molecules per cm^3 at the upper and lower vibrational levels.

²⁾ For w_i in liquids we took the probability of dissipation of the vibrational level i ,

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

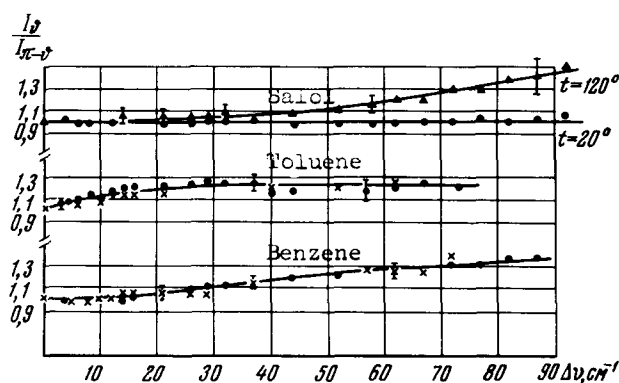
ANGULAR DISTRIBUTION OF INTENSITY IN THE THERMAL WING OF THE RAYLEIGH LINE IN LIQUIDS

G. I. Zaitsev and V. S. Starunov
 P. N. Lebedev Physics Institute, USSR Academy of Sciences
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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.