

crude model correctly captures the essence of the physics.

To get estimates of the strength of the surface potential and the magnitude of the bound-state energy, consider InAs, with a dielectric constant $\epsilon = 15$ and an effective mass $m = 0.02m_e$, where m_e is the electron mass. For a doping level of 3×10^{18} atoms/cm³, corresponding to a Fermi energy of 380 meV and $r_s = 11$, one finds $V_0 = 84$ meV and $E_b = 11$ meV.

While the direct experimental observation of these states will present considerable difficulty, largely resulting from the necessity to obtain a relatively charge-free surface, the size of the surface potential has important implications for the study of surface screening. The use of linear-response formalisms for studying this screening, even for the lowest field, is highly suspect when the zeroth-order potential is already strong enough to support a bound state. In spite of this admonition, it is nevertheless particularly interesting that the simple model for calculating the

charge distribution and potential, beginning with Eq. (9), is capable of obtaining excellent quantitative agreement with the numerical work, even though the simple model makes no use of the bound-state charge density for calculating the potential.

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Rayleigh Scattering in a Plastic Crystal Due to Orientational Relaxation

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The Rayleigh line scattered by succinonitrile is studied by means of a Fabry-Pérot interferometer having a resolving power of 5×10^5 . The line is found to be Lorentzian with a 5.4-GHz width and a depolarization ratio 0.77. The experimental data are in good agreement with the orientational relaxation theory in liquids.

At room temperature the plastic crystal of succinonitrile ($\text{N} \equiv \text{C}-\text{CH}_2-\text{CH}_2-\text{C} \equiv \text{N}$) belongs to the class $m3m$ of the cubic system.^{1,2} Several observations, especially by means of NMR,³ have shown the existence of a rotational motion of the molecule in the plastic phase. The molecule, which has a dipole moment of 3.5 D,^{4,5} is aligned along one of the ternary axes of the crystalline lattice, and the change from one orientation to another is performed by discontinuous jumps.⁶ There is also a rotational hopping motion of one of the $-\text{CH}_2-\text{C} \equiv \text{N}$ groups with respect to the other one around the axis of the molecule.⁷

A first study of the spectrum of light scattered by succinonitrile and pyvalic acid was made by

Hyde, Kevorkian, and Sherwood⁸ by means of a low-resolution interferometric spectrometer. The aim of the present work is a study of the spectral shape and the depolarization ratio of the Rayleigh line in succinonitrile.

The experimental device, a classical one, has been previously described.⁹ The resolving power of the plane Fabry-Pérot (PFP) spectrometer is equal to 5×10^5 with a free spectral range of 75 GHz. The incident light was provided by a single-frequency argon-ion laser with an output power of 80 mW for $\lambda = 4880 \text{ \AA}$. The succinonitrile single crystal, purified by zone melting, was made in a sealed cell by H. Fontaine using the Bridgman-Stockbarger technique. All the experiments were performed at 25°C. The spec-

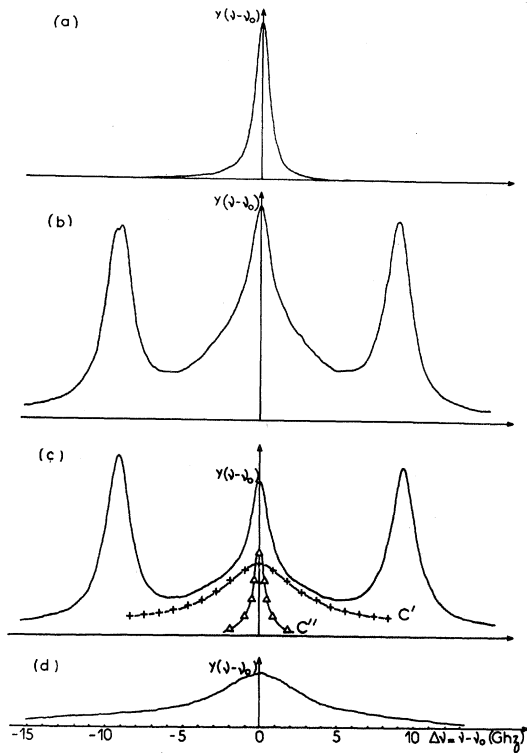


FIG. 1. (a) Instrumental profile $W(\nu-\nu_0)$. (b) Total Brillouin spectrum for succinonitrile crystal (both Vv and Vh polarizations). The scattering angle is equal to 90° . (c) Plain curve, Vv polarized spectrum; curve C' , Rayleigh line $I_{R,Vv}R_y(\nu-\nu_0)$; curve C'' , line due to elastic scattering or entropy fluctuations, $I_0W(\nu-\nu_0)$. (d) Vh polarized spectrum $I_{R,Vh}R_y(\nu-\nu_0)$.

tra have been studied in polarized light: Let V and H respectively denote the directions of polarization of the incident beam perpendicular and parallel to the scattering plane, and v and h the related directions for the scattered beam. Figure 1(a) shows the recordings of the instrumental profile, and Figs. 1(b)-1(d) the spectra for several polarization conditions of the scattered light. The recordings of Figs. 1(b) and 1(c) show that the central line is due to the superposition of several components. The broadening of Brillouin lines is noticeable on these two recordings. Its study is under investigation and will be published later. Figure 2 shows the instrumental and depolarized Rayleigh-line shapes compared with Lorentzian profiles of the same height and width. Figure 3 represents the half width at half-height of the depolarized line at several scattering angles. The dispersion of the results can be attributed to the frequency drift of the incident radiation.

The depolarization factor ρ , which is the ratio

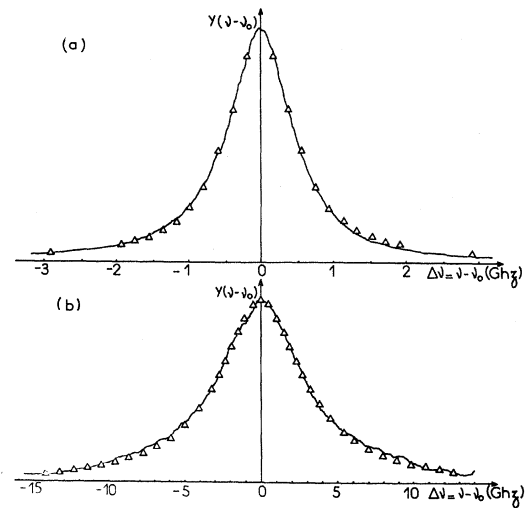


FIG. 2. (a) Instrumental and (b) Vh polarized Rayleigh profiles compared to a Lorentzian shape (triangles).

of the scattered Rayleigh powers in the Vh to that in the Vv polarization, can be identified with the ratio $I_{R,Vh}/I_{R,Vv}$ of the maximal scattered spectral densities (we assume here, as is usually done, that the spectral profile is independent of the state of polarization). These densities should be measured with accuracy in order to calculate ρ .

The recordings obtained in Vh polarization [Figs. 1(d) and 2(b)] have a Lorentzian shape, within the precision of the experiments. The density $I_{R,Vh}$ can be measured with certainty because in this state of polarization the contribution to the spectrum of any scattered light other than the inelastic Rayleigh line is negligible.

In order to measure $I_{R,Vv}$ it is necessary to eliminate the contributions of the stray light centered around ν_0 , and that of the Brillouin lines. We estimate the width of the central line using the following setup: The PFP is used as

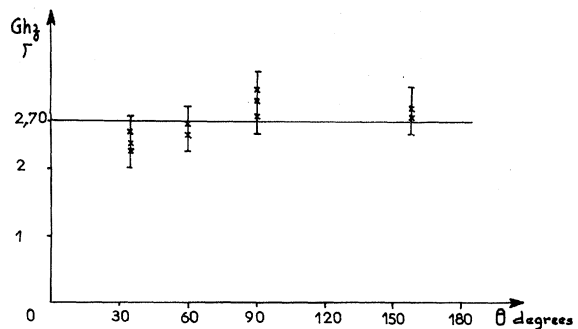


FIG. 3. Experimental values of Γ measured for several scattering angles θ .

a monochromator, and a spherical Fabry-Pérot interferometer (SFP) placed next is used to analyze the spectrum near the frequency ν_0 . The SFP has a free spectral range of 1.5 GHz and a bandwidth of 30 MHz. The resulting spectrum shows a low-level constant background due to the wide Rayleigh line, and a very narrow line centered on the frequency ν_0 having a line-width estimated to be much sharper than 30 MHz. This line, caused by the elastic scattering or by entropy fluctuations, has thus a negligible width in comparison with the PFP bandwidth (1 GHz). We represent it by the distribution $I_0\delta(\nu-\nu_0)$. Thus the recorded function $Y(\nu-\nu_0)$ [Fig. 1(c)] is given by

$$Y(\nu-\nu_0) = I_0 W(\nu-\nu_0) + I_{R,\nu_0} R_y(\nu-\nu_0) \\ + I_B B_y(\nu-\nu_{B+}) + I_B B_y(\nu-\nu_{B-}),$$

where R_y and B_y are the convolution products $R_y = R*W$ and $B_y = B*W$; the normalized functions $R(\nu-\nu_0)$ and $B(\nu-\nu_B)$ are the Rayleigh- and the Brillouin-line profiles, respectively; I_B is the maximal spectral density of the Brillouin line; and W is the instrumental transfer function.

Outside a frequency range of 10 GHz ($2\Delta\nu_0$) around ν_0 , the contribution of I_0W to Y is negligible. In the vicinity of one of the Brillouin lines the contribution of the other one is negligible too (less than 0.2% of I_B). If ν_1 and ν_2 are two frequencies symmetrical with respect to ν_{B+} or ν_{B-} and both outside the range $2\Delta\nu_0$, and if we take into account the symmetry of the Brillouin line shape, we obtain

$$Y(\nu_1-\nu_0) - Y(\nu_2-\nu_0) = I_{R,\nu_0} [R_y(\nu_1-\nu_0) - R_y(\nu_2-\nu_0)];$$

this relation allows us to calculate I_{R,ν_0} . The calculations made for many pairs ν_1, ν_2 and upon several recordings show that the dispersion of the ρ values is less than 10%.

The following results can be derived from these observations: (1) The Rayleigh line fits a Lorentzian shape with good accuracy. Half width at half-height does not depend on the scattering angle within the precision of our measurements. One finds

$$\Gamma = 2.7 \pm 0.3 \text{ GHz},$$

the corresponding relaxation time being

$$\tau = (2\pi\Gamma)^{-1} = (5.9 \pm 0.6) \times 10^{-11} \text{ sec at } 25^\circ\text{C}.$$

(2) The depolarization factor is equal to

$$\rho = 0.77 \pm 0.08.$$

To our knowledge, this is the first time that an indubitably strong inelastic Rayleigh-scattering line has been observed on a crystalline solid. Our results (strong line of large width independent of the scattering angle; depolarization factor equal to 0.77) have led us to propose the relaxation of the orientation process to account for the observed phenomena. Within the accuracy of our measurements, it turns out that the theory given by Pinnow, Candau, and Litovitz¹⁰ in the case of liquids can indeed be applied to succinonitrile. Thus, we come to the conclusion that the Rayleigh depolarized line is due to the rotation of the succinonitrile molecule in the plastic phase. Moreover, the value of τ coincides with the measured dielectric relaxation time (6×10^{-11} sec).^{4,5} The model put forward by Ivanov,¹¹ in which the molecule is assumed to keep a fixed orientation during the time τ and to jump into a different orientation in a time much smaller than τ , seems to be suitable.

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