

Measurement of the group velocity of polaritons in anthracene crystal

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The group velocity of polaritons in the range 3×10^7 – 3×10^5 cm s⁻¹ is measured from the time delay of tunable dye-laser pulses passing through thin single crystals of anthracene. A comparison of experimental results with numerical calculations gave the values of the parameters determining the dispersion curve for the lowest polariton branch.

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Measurement of the group velocity of polaritons using the time-of-flight method¹ is one of the most direct experiments that permits determining the dispersion curve of polaritons in the region of strong light-exciton mixing. Within the framework of the polariton model, the passage of light through a crystal is equivalent to the conversion of the light wave incident on the boundary of the crystal into polaritons with the same frequency, propagation of the polaritons in the crystal, and their subsequent conversion into a light wave at the opposite boundary. The time-of-flight method is based on the fact that the flight time of the polaritons in the specimen can be determined by measuring the time delay of the light pulse as it passes through the crystal and, correspondingly, the group velocity of the polaritons for known crystal thickness. Such investigations were recently performed for CuCl,² GaAs,³ and CdSe⁴ crystals. In the present work, we measured the group velocity of polaritons in anthracene crystals in the frequency range near the bottom of the lowest exciton band.

We used perfect anthracene single crystals, grown from the gas phase with a large (001) face. The measurements were performed for three specimens with thickness $d = 1.6, 11.5,$ and $35 \mu\text{m}$. The crystals were freely mounted in paper envelopes and placed in an optical cryostat in superfluid helium at $T = 1.6$ K.

Tunable dye-laser radiation was directed perpendicular to the surface of the crystal and was linearly polarized ($\mathbf{E} \parallel \mathbf{b}$ axis). In contrast to the investigations in Ref. 2–4, in which the group velocity of polaritons was measured using picosecond lasers, in our work we used a tunable dye laser with pulses of duration ~ 3 ns. The spectral width of the lasing band was $\Delta\omega_{\text{exc}} = 0.5 \text{ cm}^{-1}$ the pulse repetition frequency was 25 Hz, and the peak power was 1.5 kW. Laser radiation passing through the crystal was then directed through a DFS-12 monochromator, which was adjusted to the lasing wavelength, while the slit openings corresponded to the spectral interval $\sim \Delta\omega_{\text{exc}}$. This avoided errors in group velocity measurements in the frequency range with strong dispersion of polariton states, where the absorption coefficient increases sharply and contributions from the long wavelength wing of the lasing band can lead to considerable distortions in the shape of the laser pulse passing through the crystal.

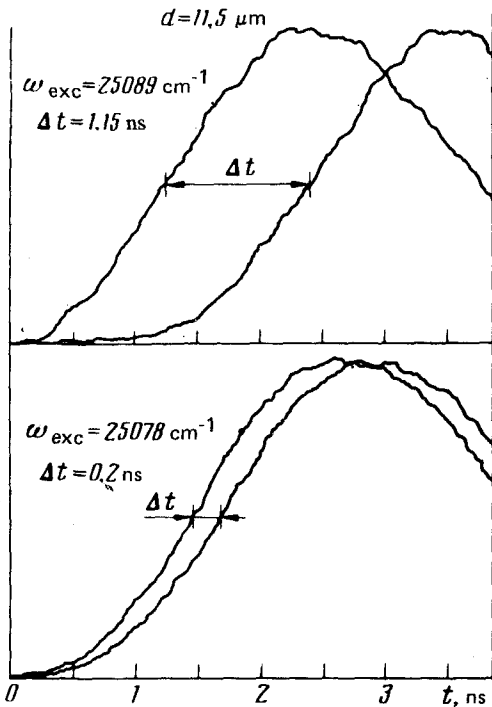


FIG. 1. Time delay Δt of a laser pulse passing through an anthracene crystal with thickness $d = 11.5 \mu\text{m}$ for two values of the frequency ω_{exc} .

The pulses were recorded at the DFS-12 outlet with the help of a 18 ÉLU-FM photomultiplier with time resolution ~ 1.2 ns, the signal from which was introduced into a S7-5 stroboscopic oscillograph. In a single measurement, ~ 2000 pulses were averaged with the help of a NTA-1024 multichannel analyzer. In the experiment, the delay time Δt of a laser pulse passing through the crystal was measured relative to an undelayed pulse with the crystal removed from the laser beam. Typical results for measurements of Δt in a crystal with thickness $d = 11.5 \mu\text{m}$ for two values of the lasing frequency ω_{exc} are shown in Fig. 1. The recording system permitted measuring the quantity Δt to within ~ 0.03 ns, which was determined by the stability of the laser pulses and the triggering stability of the measuring setup.

Figure 2 shows the measurements of the frequency dependence of the group velocity of polaritons ($V_g = d/\Delta t$), obtained in three specimens with different thicknesses.

The experimental results were compared with numerical calculations of the frequency dependence of the group velocity $V_g(\omega) = \partial\omega/\partial k$. In performing these calculations, the following relation was used for polariton dispersion:

$$\frac{c^2 k^2}{\omega^2} = \epsilon_0 + \frac{\omega_p^2 F}{(\omega_T + \frac{\hbar k^2}{2m^*} - \omega)(\omega_T + \omega)}$$

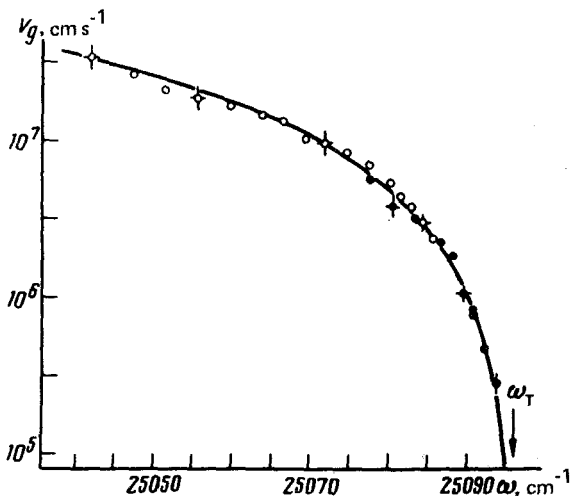


FIG. 2. Frequency dependence of the group velocity of polaritons V_g obtained in crystals with different thicknesses: \circ —35 μm , \bullet —11.5 μm , \odot —1.6 μm . The solid curve represents the calculation.

where k is the quasimomentum of the polaritons, ϵ_0 is the dielectric constant far from resonance, ω_p is the plasma frequency, F is the oscillator strength, m^* is the effective mass of the exciton, and ω_T is the frequency at the bottom of the exciton band. The values $\epsilon_0 = 2.7$ and $\omega_p^2 = 2 \times 10^8 \text{ cm}^{-2}$ were used in the calculations. The best agreement between the calculations and the experimental data was achieved with the following values of the parameters: $\omega_T = 25096 \text{ cm}^{-1}$, $F = 0.25$, $m^* = 300m_e$, where m_e is the electron mass (solid curve in Fig. 2).

It should be noted that the computed curves in $V_g(\omega)$, in the range of frequencies investigated, are most sensitive to variations in the quantity ω_T and depend to a lesser extent on the quantities F and m^* . The values of these parameters, which were determined from the experimental data, are:

$$\omega_T = 25096 \pm 0,5 \text{ cm}^{-1}, \quad F = 0.25 \pm 0.05, \quad m^* > 100 m_e.$$

In Refs. 5 and 6, following an investigation of low-temperature polariton luminescence in anthracene crystals, a number of spectral-temporal effects were explained based on the assumption that the minimum group velocity of polaritons belonging to the lowest branch V_g^{min} is lower than the velocity of sound ($u = 2-4 \times 10^5 \text{ cm s}^{-1}$ for different acoustical branches⁷). The relation $V_g^{\text{min}} < u$ is satisfied for effective exciton masses $m^* \gtrsim 100m_e$. Thus the lowest value measured in the experiment $V_g = 2.9 \times 10^5 \text{ cm s}^{-1}$, as well as the estimate $m^* > 100m_e$ obtained, are a direct confirmation of the fact that in anthracene crystals, for frequencies near ω_T , the minimum group velocity of polaritons is lower than the velocity of sound.

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