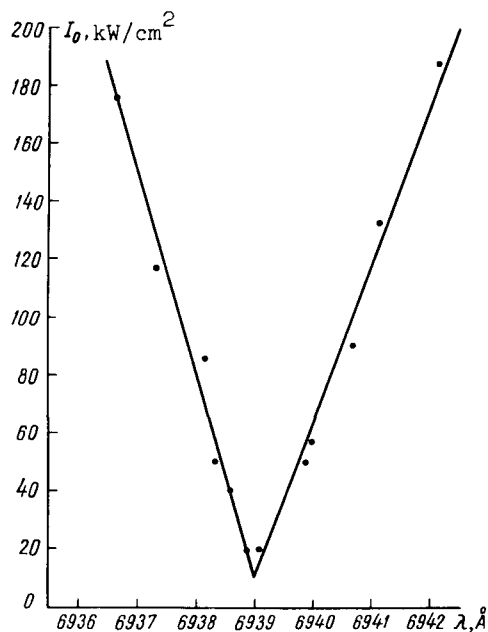


quired laser intensity on the wavelength difference $\Delta\lambda = |\lambda - \lambda_0|$ between the resonant transition and the laser emission corresponds to the theoretically expected relation (1). It follows therefore that the resonant transition $4P_{3/2} - 6S_{1/2}$ makes the main contribution to the level splitting of the $4P_{3/2}$ state of potassium under the influence of the ruby laser emission.

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Fig. 3. Ruby laser emission power density I_0 needed to obtain a signal of specified magnitude, vs. the ruby laser wavelength λ .



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1) A similar phenomenon observed in a constant electric field is reported in [3].

COLLECTIVE EFFECTS IN VIBRON SPECTRA OF MOLECULAR CRYSTALS

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One of the authors has recently shown [1] that in the interpretation of the electron-vibrational (vibron) transitions in the absorption spectra of molecular crystals sight is lost of a very important circumstance. The point is that owing to the intermolecular resonance interaction, which is well known for purely electronic excitations, virtual decay processes become possible, wherein the electronic and vibrational excitations initially localized on a single site turn out to be separated. The decay processes should produce in the region of each vibron transition broad-band two-particle absorption in which the electronic and vibrational excitons are excited simultaneously. A branch of single-particle excitations can split

off from the spectrum of the two-particle excitations, principally owing to the change $\Delta\nu$ of the vibrational frequency following the electronic excitation of the molecule. In these states the electronic and vibrational excitations move relative to each other in a limited region of space, spending part of the time on one site. The single-particle states in the absorption spectrum should correspond to narrow bands. For different oscillations, it is necessary to set in correspondence with them the known vibron K and M bands [2-3]. Two-particle absorption has not been separated as yet. Theory predicts also several singularities in the vibron spectra of molecular crystals with isotope impurities.

We have undertaken an investigation of the absorption spectra of pure and isotopic-impurity crystals of naphthalene in the region of the first vibron transition corresponding to combination with the not-fully-symmetrical oscillation B_{1g} [4].

The width of the pure-electronic exciton band E_{ex} in the naphthalene crystal is approximately $150 - 200 \text{ cm}^{-1}$ [5,6], and the change produced by the first vibron transition in the oscillation is $\Delta\nu = \nu_1 - \nu_0 = (433 - 509) = -76 \text{ cm}^{-1}$ [4] in the case of electronic excitation. Inasmuch as this yields $E_{ex} \approx 2\Delta\nu$, one should observe in the naphthalene spectrum, according to [1,5], intense two-particle absorption as well as a single-particle band preceding the two-particle absorption.

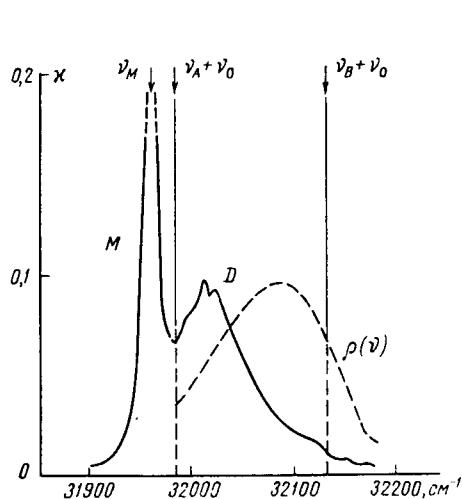


Fig. 1. Plot of the absorption of a naphthalene crystal in the region of the first vibron transition at $T = 20^\circ\text{K}$. The dashed curve shows the distribution of the state density $\rho(\nu)$ in the exciton band.

maximum of $\rho(\nu)$.

An important parameter of the one-particle states is $|a|^2$ - the quantum-mechanical probability of electronic and vibrational excitations on a single site. It is determined experimentally from the band intensities, namely: $|a|^2 = I_M / (I_M + I_D) = 0.53$. This quantity, like the distance from the M band to the edge of the D band (25 cm^{-1}), is in good agreement with

Figure 1 shows the \vec{b} -component of the absorption spectrum in the region of this vibron transition, as given by Soskin [7]. There is no doubt that the weakly-polarized M band ($\nu_M = 31960 \text{ cm}^{-1}$), for which there is practically no Davydov splitting, is a single-particle band. The same figure shows the frequencies obtained by adding the quantity ν_0 to the frequencies of the A and B bands of the pure-electronic exciton doublet. According to [1] this interval corresponds to the region of two-particle absorption. Inasmuch as the broad short-wave band accompanying the M band fits well in this interval, it can be interpreted as being a two-particle D band. The dashed curve in the figure shows the approximate distribution of the state density in the pure-electronic exciton band, $\rho(\nu)$, obtained by us from the contour of the $1 \rightarrow 0$ absorption band at $T = 90^\circ\text{K}$ [1]. In agreement with [1], the maximum of the D band is shifted to the long-wave side relative to the

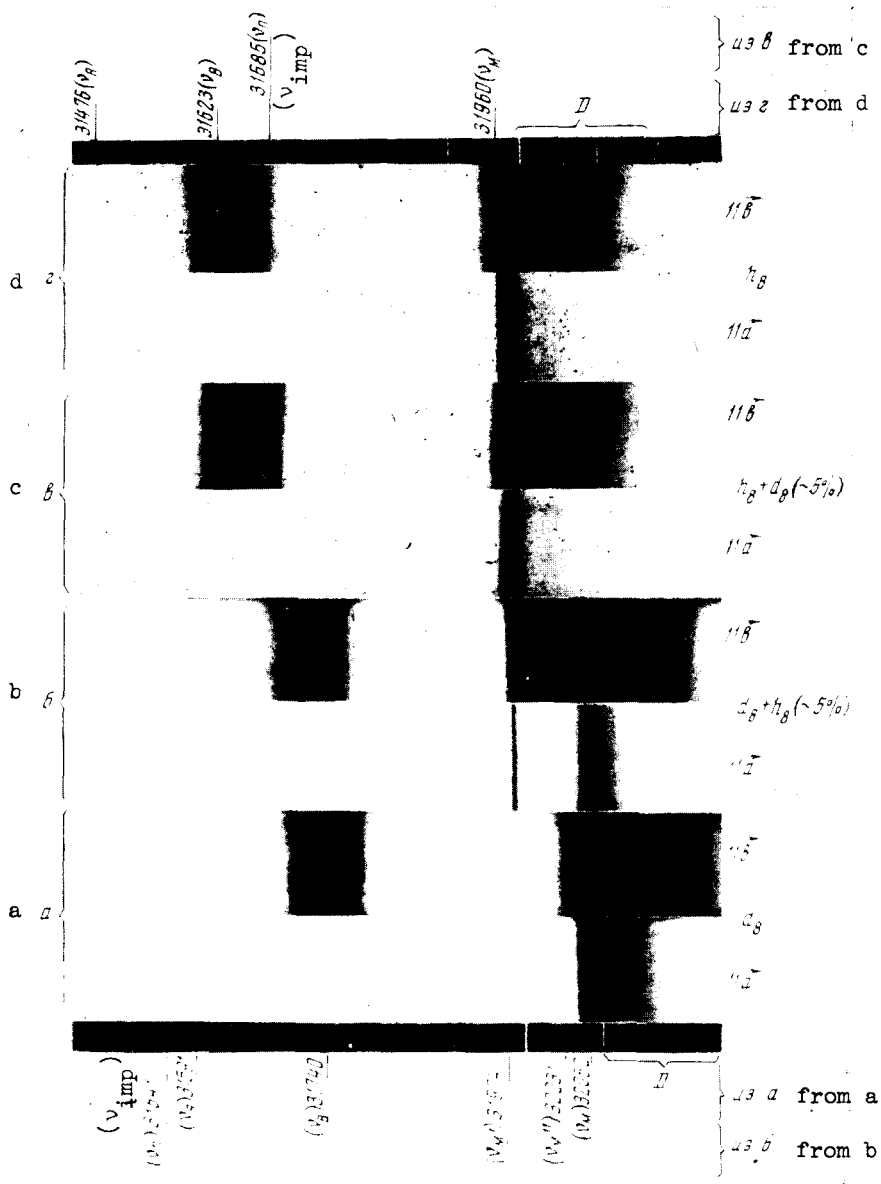


Fig. 2

the parameters of the pure electronic spectra of isotopic-impurity centers with isotopic shift $\Delta_e \approx \Delta_v$, for which the excitation is likewise only partially localized on the impurities [5, 9-11].

Important results on the connection between the observed broad-band absorption near the M band and two-particle excitations were obtained by investigating the spectra of pure and impurity crystals of paraffin h_8 and octadeuteronaphthalene d_8 in polarized light at $T = 20^\circ K$; the crystals were 2μ thick. The initial sections of these spectra are shown in Fig. 2.

Figure 2b pertains to a solution of h_8 in d_8 and corresponds to the case when the impurity spectrum lies in a region of longer wavelengths than the spectrum of the solvent (iso-

topic shift $\Delta_e < 0$). Since $\Delta_e + \Delta_v = (-115 - 76) = -191 \text{ cm}^{-1}$ is large, the usual impurity vibron band M' should appear, corresponding to excitation of vibration on the impurity molecule (we neglect the widths of the vibrational bands). In addition, according to [1], an additional vibron band M'' should appear, corresponding to the occurrence of vibrational excitation on the molecules of the solvent near the impurity. The M'' band should be smeared out and be situated between the M' and M bands, and its short-wave edge should be situated at $\nu_{\text{imp}} + \nu_0 = (31541 + 496) = 32037 \text{ cm}^{-1}$. We assume that the smeared band with maximum near $\nu \sim 32031 \text{ cm}^{-1}$ (Fig. 2b) should be set in correspondence with the M'' band. A weighty argument in favor of this is the fact that this band is not observed in the spectrum of the pure naphthalene crystal d_8 (Fig. 2a).

Figure 2c shows the spectrum of the impurity solution d_8 in h_8 , when $\Delta_e > 0$. Since here $\Delta_e + \Delta_v = (115 - 76) = 39 \text{ cm}^{-1}$, which is small compared with E_{ex} , excitation of oscillations on the impurity molecule should not give rise to a discrete level for the electronic excitation, and consequently there should be no M' band in the spectrum. It follows indeed from the spectrum of Fig. 2c that in spite of the fact that the impurity band $\nu_{\text{imp}} = 31685 \text{ cm}^{-1}$ is clearly seen in the region of the pure electronic transition, there is no narrow-band impurity absorption near the frequency 32104 cm^{-1} ($\nu_{\text{imp}} + \nu_1^{\dagger} = (31685 + 419) = 32104 \text{ cm}^{-1}$), where the M' band would be situated in the absence of decay processes. This is seen particularly clearly from the \vec{a} -component of the spectrum in Fig. 2c.

It seems to us that the data presented on the spectrum of the pure crystal, the additional vibron band M'' in the case of a low-frequency admixture, and the vanishing of the vibron band M' for the high-frequency admixture are sufficient to prove the following: (a) the strong influence of the decay processes, which are essentially collective effects, on the vibron spectrum, and (b) the presence in the vibron spectrum of two sections corresponding to single- and two-particle absorption. Direct estimates show that the fraction of the two-particle absorption in the entire vibron transition is sufficiently large and reaches 50%. This quantity demonstrates how approximate the conventional concept is, according to which the M bands are regarded as local absorption in which practically no collective effects appear.

We propose that the notion of two-particle broad-band absorption can play an important role in the interpretation of the known structure background of absorption of molecular crystals and the determination, from this interpretation, of many new parameters of exciton bands. It has been customary to avoid an analysis of this absorption, and attempts were made to exclude it from the experiment by using extremely thin crystal plates.

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1) This result is abstracted in greater detail in the paper by Davydov [8].

OSCILLATIONS OF THE PHOTOMAGNETIC EFFECT WITH THE MAGNETIC FIELD

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It might be expected on the basis of general considerations that the magnitude of photomagnetic effects in semiconductors should oscillate under certain conditions with the magnetic field. Recently, at the suggestion of Yu. M. Kagan, a theory was developed [1], from which it follows that oscillations of the photomagnetic effect should be observed in relatively weak magnetic fields if the following conditions are satisfied: the carrier relaxation time is $\tau \gg m^*c/eH \equiv 1/\omega$ (m^* = carrier effective mass, H = magnetic field intensity, c = velocity of light, e = electron charge, ω = carrier cyclotron frequency), and the Fermi energy is $E_F \gg \hbar\omega \gg kT$. These conditions are satisfied by indium antimonide at low temperatures. We have therefore undertaken an investigation of both the even and odd photomagnetic effects in InSb in magnetic fields up to 23,000 Oe at liquid-helium temperature ($T = 4.2^\circ\text{K}$). The carrier density in the investigated sample was 10^{15} at/cm³. The sample was equipped with two electrodes for the measurement of the odd photomagnetic emf, and two electrodes perpendicular to them to measure the even photomagnetic emf. During the investigation of the even photomagnetic effect the plane of the sample was inclined 30° to the direction of the magnetic field. Measurements of the even photomagnetic emf were made in the direction of the projection of the magnetic field on the plane of the sample [2].

The figure shows the odd and even photomagnetic emf's (curves 1 and 2, respectively), as functions of

