Rashba effect in a hydrostatically compressed crystal of deuteronaphthalene

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An experimental investigation was made of the Rashba effect in a doped crystal of deutero-naphthalene subjected to hydrostatic pressures up to 15 kbar. The absorption spectra at various pressures were determined and it was found that an increase in the pressure shifted the exciton and impurity absorption bands in the direction of the red wavelengths, increased the Davydov splitting, and lowered the exciton-impurity binding energy ε. The dependence of ε on the width of the exciton spectrum was determined and it was established that a quasiresonance between an impurity-molecule level and the level of the host crystal molecules increased on increase in the width of the exciton spectrum. The vibronic-luminescence spectra were recorded and the dependence of the amplitude $|a_0|^2$ of localization of an exciton at an impurity on the binding energy ε was determined in a wide range. A reduction in ε resulted in a progressive delocalization of the impurity excitation. The Rashba-theory formulas were used to calculate the dependences of ε and $|a_0|^2$ on the width of the exciton spectrum, and a comparison was made with the experimental results. These experimental results were described well by Rashba's theory.

The Rashba effect or a quasiresonance between a level of an impurity molecule and the levels of the host crystal was first discovered and investigated for isotopic-impurity deuteronaphthalene crystals.[1-3] The physical essence of the effect is a strong dependence of the impurity absorption on the gap Δ separating the energy levels of the solvent (host crystal) and impurity molecules, and on the strength m of the intermolecular resonance interaction that governs the width of exciton energy bands. If $\Delta \gg m$, the impurity-absorption intensity is governed by the dipole moment of a transition in an isolated molecule, whereas the polarization ratio is determined by the orientation of this dipole relative to the crystallographic axes (this is known as the oriented-gas model). However, if $\Delta \approx m$, an impurity excitation is no longer localized at an impurity molecule but extends to the nearby molecules of the solvent (host crystal), which affect strongly the impurity absorption. Finally, if $\Delta \lesssim m$, then the excitation is largely concentrated at the solvent molecules and extends over a region considerably greater than the lattice period. In this case the quantitative characteristics of the impurity absorption are no longer described by the oriented-gas model, but are governed mainly by an exciton-energy band in the crystal; this provides an opportunity for investigating the structure of such a band and particularly the positions of its edges.[4]

The traditional use of isotopic-impurity deuteronaphthalene crystals for the investigation of this quasiresonance is due to extremely favorable properties of these crystals. In fact, isotopic substitution gives rise to differences Δ ranging from -118 to $44\,\mathrm{cm}^{-1}$ between the electron excitation energies of the isotopic forms of naphthalene, whereas the resonance interaction is practically unaffected. The quantitative parameter used to describe the degree of localization of an excitation at an impurity is the ratio Δ/m, which in the case of isotopic-impurity deuteronaphthalene crystals ranges from 1.3 to 0.1. This has made these crystals the most popular objects for quantitative investigations of

the characteristics of the impurity absorption as a function of the value of Δ/m (Refs. 5 and 6). However, it should be pointed out that these crystals suffer also from a number of disadvantages. They include, above all, the limited number of the deuteroderivatives of naphthalene and the consequent discrete variation of the parameter Δ/m. Considerable errors in the measurements result from the presence of accidental deuteroimpurities which are unavoidably present in isotopic-impurity crystals. Finally, in quantitative studies we have to allow also for the difference between the spectra of the individual deuteroderivatives composing an isotopic-impurity crystal and varying on transition from one crystal to another.

A new approach to a study of the Rashba effect is currently being developed: it is based on the use of high-pressure optical chambers in low-temperature spectroscopic measurements. It is shown in Ref. 7 that hydrostatic compression of naphthalene crystals shifts the exciton and impurity absorption bands; such compression increases rapidly the Davydov splitting of the bands which determines the width of the exciton spectrum. This makes it possible to control the parameter Δ/m by varying it in a wide range practically continuously and permits a quantitative investigation of the characteristics of impurity absorption under quasiresonance conditions in the same crystal containing the isotopic impurity. This approach is also attractive because it allows us to study the Rashba effect of deep impurities with a strong localization of the excitation and to investigate experimentally the opposite limiting case of weak localization of an excitation at an impurity right up to a situation of complete delocalization.

We carried out an investigation of the Rashba effect in a hydrostatically compressed deuteronaphthalene crystal containing protonated naphthalene and β-methylnaphthalene impurities. We determined the absorption and luminescence spectra in a wide range of pressures and then used these spectra to find the dependence of the binding energy ε

and of the localization amplitude $|a_0|^2$ of an exciton at an impurity on the parameter Δ/m. We calculated these quantitites from Rashba's theory formulas using the experimentally found density of the exciton states[4] and we compared these calculations with our experimental results.

1. EXPERIMENTAL METHOD

The absorption and luminescence spectra were recorded at $T = 4.2$ K using unpolarized light and a spectrometric system based on a DFS 13-2 spectrograph (dispersion 2 Å/mm; relative aperture 1:40) with a photoelectric attachment of the FÉP-2 type. The radiation detector was an FÉU-106 photomultiplier. The luminescence spectra were excited by a kilowatt DKSÉL-1000 xenon lamp with a stabilized light output. The part of the spectrum necessary for the excitation of the luminescence was selected by a high-luminosity monochromator of the MS type (dispersion 13 Å/mm; relative aperture 1:2.5). The absorption spectra were recorded employing a 200-W DKSSh-200 xenon lamp emitting white radiation. Overheating of a sample was prevented by removing the thermal radiation of this lamp with a water filter. The spectral slit width was 0.3 Å in all the measurements. The control of the spectrometer and of its output, as well as acquisition, storage, and primary analysis of the spectra were performed by a microcomputer with a module interface which provided a bidirectional coupling to a more advanced computer. The microcomputer also analyzed the experimental transmission spectra and determined the spectral dependences of the extinction coefficient. A helium optical cryostat with quartz windows was used in low-temperature measurements. It was provided with an automatic temperature stabilization system which kept the temperature constant to within 10^{-2} K in the range 4.2–300 K. The investigated crystals were plates 10–100 μ thick with a highly developed (ab) plane and were grown by sublimation in a nitrogen atmosphere from a material first purified by zone melting. The naphthalene impurity was introduced by adding a suitable amount to the initial charge and its concentration, deduced from the absorption spectra, did not exceed 0.5%. The concentration of β-methylnaphthalene, estimated by the same method, did not exceed 0.05%.

Crystals were compressed using a high-pressure chamber with leucosapphire anvils which also acted as optical windows. Leakage of the pressure-transmitting medium and damage to the anvils at the points of maximum compression were prevented by using a sealing gasket made of cold-rolled beryllium bronze. A sample was placed at the center of the gasket in a recess 1.2–0.95 mm in diameter and 0.4 mm deep. The anvils were driven by a minipress which provided a usable force of about 0.5 metric tons. The construction of the press made it possible to use a standard helium optical cryostat with a diameter 32 mm in the low-temperature measurements; the optical windows of the chamber and thermostat were combined. A detailed description of the high-pressure chamber was given in Ref. 8. The pressure in the chamber was measured by the method of optical calibration.[9,10] The pressure was deduced from a shift of the R_1 luminescence line of a ruby crystal of about 100 μ size,

placed inside the chamber alongside the investigated sample.

The error in the determination of pressure from such a ruby luminescence scale was governed by the precision with which the spectral shift of the R_1 line was determined. The use of a high-resolution spectrometer made it possible to find this line shift to within 10^{-2} nm; the error in the determination of the pressure was then about 0.3 kbar. The investigated naphthalene crystal had a diameter of about 900 μ and it was cut from a plate 20–50 μ thick; it was placed inside the gasket recess in such a way it did not touch the walls of the recess or the ruby crystal. Next, the pressure-transmitting liquid was poured into the recess and (after removal of air bubbles) the anvils were loaded by the minipress. The pressure-transmitting medium was a mixture of 25% alcohol and 75% glycerin. The line broadening was minimized by loading to the required pressure at room temperature when the pressure-transmitting medium was liquid and the hydrostatic compression was ideal. Next, the chamber was cooled slowly to the working temperature $T = 4.2$ K. This made it possible to record high-quality absorption and luminescence spectra in which an inhomogeneous line broadening (compared with a free crystal) did not exceed 3 cm^{-1} when the absolute shift was 300 cm^{-1}.

2. EXPERIMENTAL RESULTS

Figure 1 shows the absorption spectra of a doped deuteronaphthalene crystal, recorded in the region of the lowest singlet exciton transition at 4.2 K and various pressures. At atmospheric pressure the spectrum consisted of a narrow A band ($\omega = 31\,588$ cm^{-1}, A_u representation, factor group C_{2h}) and a wide B band ($\omega = 31\,739$ cm^{-1}, B_u representation), polarized respectively perpendicular and parallel to the monoclinic crystal axis and forming an exciton Davydov doublet.[11,12] The A band of this exciton doublet corresponded to the bottom of the exciton energy band of the host crystal, forming a continuous spectrum of 180 cm^{-1} width.
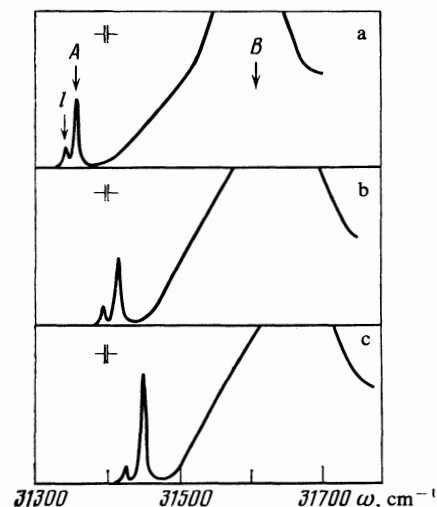

FIG. 1. Exciton absorption spectra of isotopic impurity crystals containing 0.5% of $C_{10}H_8$ in $C_{10}D_8$ at $T = 4.2$ K, recorded under various pressures: a) 14.6 kbar; b) 12.96 kbar; c) 10.8 kbar. The long-wavelength peak (I) is the absorption band of the impurity; A and B are the bands forming an exciton doublet.

The upper edge of the exciton energy band was located 30 cm^{-1} above the maximum of the B absorption band in the exciton doublet.[4] The spectra were recorded using unpolarized light because the anvils depolarized the incident light; therefore, the two bands were present simultaneously in the spectrum. The absorption band of the naphthalene impurity was located at 49 cm^{-1} below the A band ($\omega_{00} = 31\ 539$ cm^{-1}). The separation from the impurity band to the unsplit molecular term[1)] of deuteronaphthalene was 124 cm^{-1}, which was greater than the isotopic shift (118 cm^{-1}) between the electronic levels of these molecules in the vapor phase. It should be pointed out that this was a consequence of a quasiresonance between the impurity level and the levels of the molecules in the host crystal, because the shift of the electronic levels as a result of the vapor-crystal transition and due to the influence of the crystal field was the same for both molecules.[13]

It is clear from Fig. 1 that an increase in the pressure shifted all the bands to the red part of the spectrum; the Davydov splitting of the exciton absorption bands increased and the separation between the impurity and A bands decreased. It was shown by us earlier[7] that the rates of shift of the exciton absorption bands were different, due to the difference between the nature of the intermolecular interaction forces. The relative shift of the absorption bands, defined as the ratio of the vapor-crystal shifts under pressure and in absence of pressure, for the impurity and the unsplit molecular term was due to the dispersive van der Waals forces and depended quadratically on the relative change in the volume V_0/V_p. The relative value of the Davydov splitting of the exciton absorption bands found in this way was governed by the resonance interaction and, as demonstrated by us experimentally on an earlier occasion,[7] it was proportional to the fourth power of the relative change in the volume V_0/V_p. Since the vapor-crystal shift was the same for the naphthalene and deuteronaphthalene molecules, the rates of shift of the impurity band of the unsplit molecular term should also be the same in the absence of a quasiresonance.

In the case of the exciton absorption bands the resonance interaction increased the rate of shift of the A band and reduced the rate of shift of the B band, compared with the unsplit molecular term. The experimentally determined average rates of shifts of the absorption bands were 12.5, 14.9, and 7.9 cm^{-1}/kbar, respectively, for the naphthalene impurity and for the A and B bands of the exciton doublet. Therefore, an increase in the pressure resulted in a gradual approach of the impurity band of naphthalene to the A band of the exciton absorption doublet until they overlapped. A strong pressure dependence of the Davydov splitting made it possible to study the region of the overlap of the impurity and A bands (crossover region) at relatively low pressures which were attainable in chambers with sapphire anvils.

Figure 2a shows the pressure dependences of the positions of the impurity and exciton absorption bands of the investigated crystal. The precision of determination of the positions of the impurity and A bands was 1 cm^{-1}. In the case of the B band the error in the determination of its center of gravity was approximately 3–4 cm^{-1}, because the intensity of this band and its width were greater. It is clear from this figure that at pressures above 15 kbar the separation between the impurity band and the A band of the doublet decreased to 10 cm^{-1}. However, a direct investigation of the crossover region met with difficulties encountered in maintenance of the high pressure, which arose from the specific construction of the chamber. The slow relaxation of the pressure after compression broadened the lines and made it difficult to separate the impurity band from the A band of the exciton doublet. In the lower part of the figure we plotted the dependence of the position of the impurity band of β-methylnaphthalene deduced from the luminescence spectra. The rate of shift of the β-methylnaphthalene band was 12.4 cm^{-1}/kbar.

The pressure dependences of the positions of the absorption bands deduced from the spectra could be used to find the dependence of the energy ε representing the binding of an exciton to an impurity on the Davydov splitting. The Rashba effect theory made it possible to analyze quantitatively this dependence (the results will be given in the next section). The experimental dependence is shown in Fig. 2b. The straight line in this figure is the same dependence in the absence of a quasiresonance when an excitation is localized entirely at an impurity, which corresponds to the oriented gas model. We can see from the figure that repulsion of the impurity level, which increases on approach to the bottom of the exciton energy band, occurs under quasiresonance con-
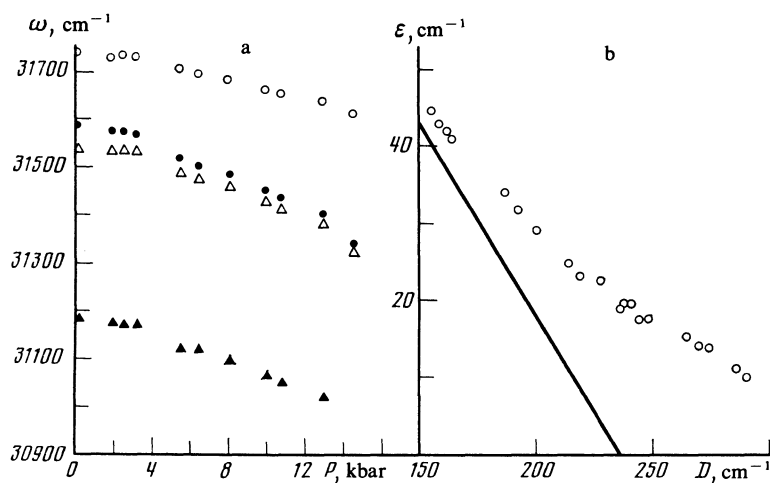

FIG. 2. a) Pressure dependences of the positions of the absorption bands of an isotopic-impurity crystal with 0.5% of $C_{10}H_8$ in $C_{10}D_8$: O) B band; ●) A band; △) $C_{10}H_8$ impurity; ▲) β-methylnapthalene impurity. b) Dependence of the energy ε representing the binding of an exciton to an impurity on the Davydov splitting D. The stright line is based on the oriented gas model.

ditions.

An important parameter, which changes greatly under quasiresonance conditions and describes directly the delocalization of an impurity excitation is the amplitude of localization of the excitation at an impurity position. An experimental method for the determination of this parameter was put forward by Broude et al.[14] The method is based on the observation that deuteration of molecules alters the frequency of some intramolecular vibrations by a few percent. Under quasiresonance conditions, when an impurity excitation is delocalized and extendes to the nearby molecules of the host crystal, an electron-phonon spectral band corresponding to transitions involving the same vibration (which has different frequencies for the impurity molecule and the host crystal molecule) becomes a doublet. One of the components of this doublet represents a transition from an impurity level to an intramolecular vibration of the impurity, whereas the other component corresponds to a transition from the same level to an intramolecular vibration of the host crystal. The ratio of the intensities of the impurity component to the combined intensity of both components of the doublet determines the amplitude $|a_0|^2$ of localization of an excitation at an impurity. In the case of a naphthalene crystal it is convenient to find $|a_0|^2$ from a strong electron-phonon transition involving an intramolecular partly symmetric vibration at $\nu = 509$ cm^{-1}, which has the B_{3g} symmetry and which occurs at $\nu = 493$ cm^{-1} in the case of deuteronaphthalene. In the spectrum of a deuteronaphthalene crystal doped with naphthalene this band splits into two components separated by 16 cm^{-1}. At atmospheric pressure the localization amplitude was $|a_0|^2 = 0.83$, i.e., the impurity component predominated. On approach of the impurity level to the exciton energy band the quasiresonance changed to a progressive delocalization of the impurity excitation and this should result in a redistribution of the intensity between the components of the doublet. Figure 3 shows the luminescence spectra of a doped deuteronaphthalene crystal recorded in the

FIG. 3. Vibronic luminescence spectra of an isotopic-impurity crystal with 0.5% of $C_{10}H_8$ in $C_{10}D_8$, recorded at $T = 4.2$ K at pressures of $p = 12.97$ kbar (1), 10.8 kbar (2), and 8.05 kbar (3). The inset shows energy level scheme of isotopic-impurity complexes: the dashed arrow is the short-wavelength component of a doublet involving a crystal vibration at $\nu = 493$ cm^{-1}; the continuous arrow represents the long-wavelength component of a doublet with participation of an impurity vibration at $\nu = 509$ cm^{-1}.

range corresponding to the electron-phonon transitions involving the $\nu = 509$ cm^{-1} (493 cm^{-1}) vibrations at the temperature of 4.2 K and under various pressures. The inset in this figure gives an energy level scheme for this crystal, where the transitions corresponding to the doublet components are identified by arrows. The continuous arrow is the long-wavelength component of the doublet, corresponding to a transition from an impurity level to an intramolecular vibration of the impurity, whereas the dashed arrow represents a short-wavelength component corresponding to a transition from the same level but to an intramolecular vibration of the host crystal. The separation between the doublet components was found to be 16 cm^{-1} at all pressures and the position of the impurity component shifted relative to the purely electronic resonance absorption band of the impurity at 509 cm^{-1}. It is clear from Fig. 3 that an increase in the pressure and approach of the impurity level to the bottom of the exciton energy band enhanced the low-wavelength component of the doublet. Hence, a reduction in the binding energy of an exciton attached to an impurity reduced also the amplitude of localization of the exciton $|a_0|^2$, i.e., under quasiresonance conditions an impurity excitation was partly delocalized. Since the broadening of the spectral bands in experiments under hydrostatic pressures did not exceed 3–4 cm^{-1}, the bands in the doublet were well resolved and the experimental spectra could be used to determine accurately the dependence of $|a_0|^2$ on ε to make a quantitative comparison with the theory.

3. DISCUSSION OF RESULTS

A quantitative analysis of the experimental results was made on the basis of the theory of absorption of light by local excitons developed by Rashba for an isotopic impurity, when the individual impurity centers are isolated and do not interact with one another.[1–3,15] This theory was founded on a theory of degenerate perturbations developed for solids first by Lifshitz,[16] and then by Koster and Slater.[17] The theory analyses the integrated intensity and polarization of the impurity absorption bands, positions of these bands, amplitude of localization of excitations at the impurities, and impurity-induced absorption in the exciton continuum. In a quantitative analysis of our experimental results we calculated the positions of the impurity absorption bands and the amplitudes of localization of excitations at impurites on the basis of Rashba's theory.

Positions of impurity absorption bands. If the isotopic shift Δ is sufficiently large, a discrete level appears and it corresponds to the formation of a local exciton. The position ε of a discrete level of a local exciton is governed by an isolated pole of the Green function of a pure crystal:

$$\Delta G_0(\varepsilon) = 1. \tag{1}$$

Here

$$G_0(\varepsilon) = \int \frac{\rho(\nu)}{\varepsilon - \nu} d\nu$$

is the Green function of a pure crystal; $\rho(\nu)$ is the density of exciton states in the crystal; integration with respect to ν is carried out throughout the range where $\rho(\nu) \neq 0$. The root ε
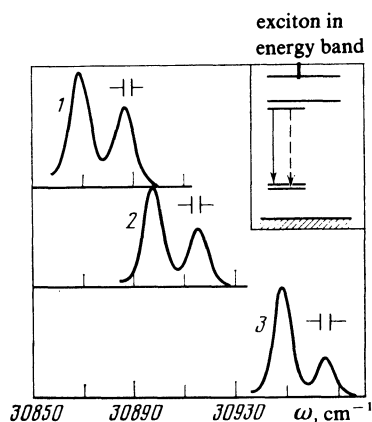
of Eq. (1) which gives the position of the impurity level is outside the exciton spectrum where $\rho(\nu) = 0$.

The amplitude of localization of an exciton at an impurity molecule is given by the following expression:

$$|a_0(\varepsilon)|^2 = \frac{d\varepsilon}{d\Delta} = \left[\frac{d}{d\varepsilon}\frac{1}{G_0(\varepsilon)}\right]^{-1} = \frac{G_0^2(\varepsilon)}{dG_0(\varepsilon)/d\varepsilon}. \quad (2)$$

Therefore, we can calculate the positions of the impurity bands, the energy level ε, and the amplitude of localization of an exciton at an impurity $|a_0(\varepsilon)|^2$ if we know the density of the exciton states $\rho(\nu)$ and the isotopic shift Δ. In the hydrostatic compression experiments the isotopic shift is constant and equal to 118 cm^{-1}, whereas the width of the exciton spectrum increases on compression. The density of the exciton states $\rho(\nu)$ in a naphthalene crystal, deduced experimentally from the profiles of the vibronic band-band transitions in the luminescence spectrum allowing for the positions of the impurity absorption bands under the Rashba effect conditions, is known from Ref. 4. However, in order to calculate ε and $|a_0|^2$, we have to know how they vary under hydrostatic compression. Since there was no experimental data, we shall assume that the nature of the distribution of the density states is not affected by hydrostatic compression, but broadens proportionally on the energy scale. The coefficient of proportionality can be found from the experimental value of the Davydov splitting of exciton bands at a given pressure. To some extent this hypothesis is supported by the results of Ref. 18, where plots of phonon dispersion in a naphthalene crystal under pressure were given and it was shown that their profile was not affected. The calculations were made on a computer. The error of these calculations was reduced by dividing the integration interval in the formula for G_0 into 2000 points, and the results of integration and subsequent differentiation were smoothed out by second-degree polynomials.

Figure 4 gives the experimental (points) and calculated (dashed curve) dependences of the positions of impurity bands on the relative change in the Davydov splitting D_p / D_0. We can see from this figure that the experimental points fit well the calculated curve. It should be pointed out that the
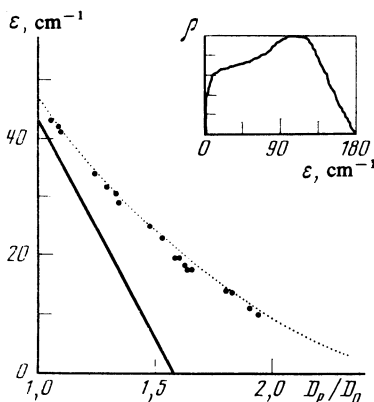

FIG. 4. Exciton-impurity binding energy ε versus the relative value of the Davydov splitting D_p/D_0. The points are the experimental values and the dotted curve is calculated; the continuous line is based on the oriented gas model. The inset shows the density of the exciton states in a crystal.
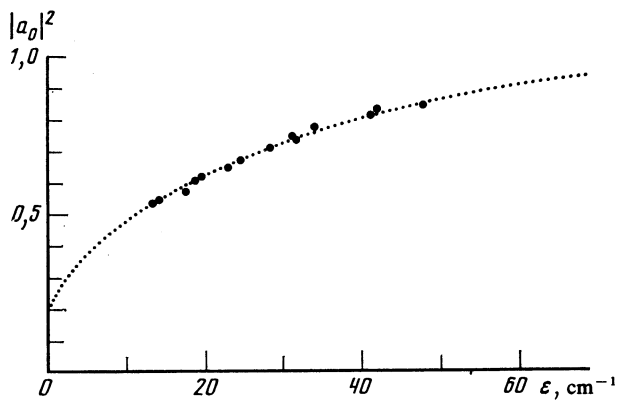

FIG. 5. Dependence of the amplitude of localization of an exciton at an impurity on the exciton binding energy ε. The points are the experimental results and the dotted line is calculated.

experimental error in the determination of ε does not exceed 0.5 cm^{-1}, but it is large in the case of the Davydov splitting of the bands when it is governed by the error in the determination of the position of the B band amounting to 3–4 cm^{-1}. The continuous curve in Fig. 4 shows this dependence deduced on the basis of the oriented-gas model. Figure 5 gives the experimental (points) and calculated (dotted curve) dependences of the amplitude $|a_0|^2$ of the localization of an exciton at an impurity on the exciton binding energy ε. It is clear from Fig. 5 that the experimental points fit excellently the calculated curve and the precision of the measurements is high. In the absence of a quasiresonance an exciton is localized completely at an impurity and we have $|a_0|^2 = 1$ irrespective of the value of ε.

The experimental data on the shift of the band of β-methylnaphthalene cannot be interpreted as a manifestation of a quasiresonance. In fact, the rate of shift of the bands, governed primarily by dispersive forces, depends on the vapor-crystal shift, which in the case of β-methylnaphthalene is approximately 10% greater than in the case of naphthalene. This accounts for the greater (compared with the unsplit molecular term of naphthalene) shift of the β-methylnaphthalene impurity bands. However, estimates show that the possible shift due to a quasiresonance is considerably less than that found experimentally. It follows from this account that it is difficult to observe a quasiresonance using the band shift of chemical impurities and in this case it is necessary to employ other methods, such as determination of the polarization of the absorption bands.

Our experimental results demonstrate convincingly that a quasiresonance occurs when an impurity level approaches the edge of the exciton energy spectrum under hydrostatic compression conditions, and the results are in quantitative agreement with calculations carried out using the Rashba theory formulas. This provides an additional experimental proof of the correctness of the quasiresonance theory and possibly accounts for the experimentally determined density of the exciton states in a naphthalene crystal and supports the hypothesis on the nature of changes in this density under hydrostatic pressures. However, it should be pointed out that the experimental difficulties prevented us

from investigating the most interesting and so far unknown crossover region. An improvement in the high-pressure chamber should make it possible to solve this problem. It should also be pointed out that the method of hydrostatic compression, which can be used to alter the energy spectrum of molecular crystals, provides new opportunities for a quantitative investigation of the effects of a resonance interaction and the laws governing the vibronic absorption of light.

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[1)]An unsplit molecular term determines the position of a purely electronic absorption band of a crystal without a resonance interaction, which would result in Davydov splitting and give rise to an exciton energy band in crystals with several molecules per unit cell. In the case of a naphthalene crystal, this term is located half-way between the Davydov doublet bands.[13]

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