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*DIAMAGNETIC EXCITONS IN SEMICONDUCTORS**

B. P. ZAKHARCHENYA and R. P. SEĬSYAN

A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences

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THIS paper is concerned more with the problem of magneto-optical investigations of semiconductors than with exciton states. We shall consider one of the fundamental aspects of this problem, namely, that of allowing for the Coulomb interaction in interband optical transitions in a semiconductor subjected to a magnetic field.

It is well known that Landau sub-bands appear in the valence and conduction bands of a crystal in sufficiently strong magnetic fields; the separation between these sub-bands is, neglecting spin, equal to $\hbar\omega_c$, where $\omega_c = eH/m^*c$ is the frequency of orbital motion (the cyclotron frequency) of electrons of mass m_e^* or holes with a mass m_h^*. The orbital motion of carriers takes place in a plane perpendicular to the magnetic field and carrier states are similar to quantum states of a harmonic oscillator with energies $\mathcal{E} = (l + 1/2)\hbar\omega_c$, while the motion along the magnetic field is infinite (carriers move along a helical path) and is represented by the projection of a wave vector k_z onto the direction of the magnetic field. Using this model of the interaction of free carriers in a semiconductor with the magnetic field, we can speak of one-dimensional Landau sub-bands in a crystal.

Optical interband transitions, taking place between systems of such sub-bands, each of which is represented by a quantum number $l = 0, 1, 2, \ldots$, should give rise to a discrete structure of the edge of continuous absorption of a crystal (Fig. 1). A magnetic field regroups the density of states in a three-dimensional band into one-dimensional sub-bands. The density of states then increases to infinity at the sub-band edges. In this case, the absorption coefficient is proportional to $(\hbar\omega - \mathcal{E}_{ll'})^{-1/2}$, where $\mathcal{E}_{ll'} = \mathcal{E}_g + (l + 1/2)\hbar\omega_c^e + (l' + 1/2)\hbar\omega_c^h$. The absorption coefficient should become infinite when $\hbar\omega = \mathcal{E}_{ll'}$ (transitions take place between the sub-band edges) and should decrease rapidly away from the edges (Fig. 1b). Thus, we should expect the absorption spectrum to have periodically repeated narrow asymmetric maxima, with an infinitely high value of the abosrption coefficient at each maximum. However, the experimentally observed absorption maxima are always finite (cf., for example, Fig. 9). This serious disagreement between the experiment and theory is usually removed by allowing for nonstationary effects, which may be due to scattering by phonons or charged impurities.

The appearance of such a discrete structure at the absorption edge is usually called the magnetoabsorption oscillation effect. Just as the Zeeman effect is regarded as the principal magneto-optical phenomenon in the case of free atoms, the magnetoabsorption oscillations can be regarded as the principal effect in the magneto-optics of semiconductors. In fact, several "derivatives" are associated with this principal phenomenon, including the interband Faraday effect, the Voigt effect (birefringence in a magnetic field), magnetoabsorption in crossed electric and magnetic fields, and other phenomena.

Analysis of the structure of the magnetoabsorption spectrum is always specific to a given case and makes it possible to obtain extensive information on the band structure of a semiconductor, to calculate its band-structure parameters with a high accuracy, to determine the nature of the interband transitions, and, in particular, to find from the nature of the spectrum whether these transitions are direct allowed, forbidden, or indirect.

The magnetoabsorption oscillation effect was discovered in 1957 by three groups of investigators, working independently on different semiconductors: Zwerdling and Lax found it in germanium,[1] Burstein and Picus discovered it in indium antimonide,[2] Gross, Zakharchenya, and Pavinskiĭ observed it in cuprous oxide.[3] At present, the number of semiconducting compounds in which this phenomenon was found is close to twenty. The technique of investigating the effect has been modernized. For example, details of the effect are being investigated using sensitive methods of electromagneto- and piezomagneto-absorption and reflection. Static magnetic fields of intensities higher than 100 kOe are used in the USA at the Francis Bitter National Magnetic Laboratory and at the Naval Research Laboratories, Washington.

In spite of the considerable progress in investigations of the magnetoabsorption oscillations, there re-

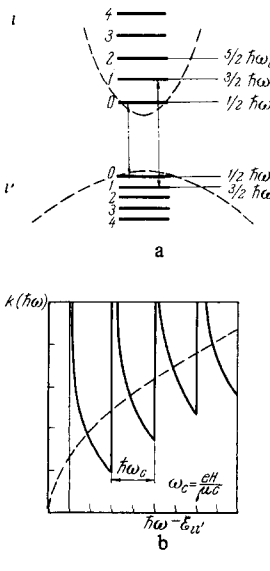

FIG. 1. a) Optical transitions between systems of Landau sub-bands in a semiconductor. The dashed parabolic curves represent the valence and conduction bands in H = 0. b) Absorption coefficient as a function of $\hbar\omega - \mathcal{E}_{ll'}$, where $\mathcal{E}_{ll'} = \mathcal{E}_g + (l + 1/2)\hbar\omega_c^e + (l' + 1/2)\hbar\omega_c^h$. The dashed curve shows a continuous dependence $k(\hbar\omega)$ in the absence of a field, when $\mathcal{E}_{ll'} = \mathcal{E}_g$, which is the energy gap between the valence and conduction bands. The solid curve shows the discontinuous nature of the absorption coefficients between the Landau sub-bands when $k \propto (\hbar\omega - \mathcal{E}_{ll'})^{-1}$ and the scattering by free carriers is ignored.

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mained until recently the largely unresolved problem of the participation of exciton states in the interband magneto-optical transitions, which is important for the understanding of the true nature of the phenomenon. There had been, however, in one of the first papers reporting the discovery of the effect, the suggestion that the discrete structure appearing in a magnetic field beyond the continuous absorption edge was not due to optical transitions taking place directly between the Landau sub-band edges but was the consequence of transitions to exciton states associated with each pair (hole and electron) of Landau sub-bands which combine to produce an optical transition (Fig. 2). Such excitons may be called arbitrarily the diamagnetic excitons in view of their relationship with the diamagnetic Landau sub-bands.

One of the first attempts to make a theoretical allowance for the influence of the Coulomb interaction on the nature of absorption in a strong magnetic field was made by Bulyanitsa and Pavinskiĭ.[4] Yafet, Keyes, and Adams[5] derived a wave function for the ground state of a hydrogen-like system with a reduced mass $\mu = m_e^* m_h^*/(m_e^* + m_h^*)$, which is moving orbitally in a medium of permittivity κ in a strong magnetic field.

The best known work dealing with the problem considered here is that of Elliott and Loudon.[6] Solutions of an equation, written in the effective mass approximation for the orbital motion in a magnetic field:

$$\left[\frac{1}{2\mu}\left(\mathbf{p}+\frac{e}{2c}[\mathrm{H}\times\mathrm{r}]\right)^2-\frac{e^2}{\kappa r}\right]\Psi(r)\mathscr{E}=\Psi(r), \tag{1}$$

are sought on the assumption of the following relationship

$$\beta=\frac{\kappa^2\hbar^3 H}{\mu^2 e^3 c}\gg 1, \tag{2}$$

which defines the strong field condition. We can easily see that 2β is the ratio of the energy of the cyclotron motion to the binding energy of an exciton, which—for the ground state—is equal to the Rydberg constant for an exciton $R_{ex} = \mu e^4/2\hbar^2\kappa^2$. Thus, Eq. (2) is equivalent to the statement that the strong field approximation is satisfied when the separation between the Landau sub-bands becomes much greater than the exciton energy in the absence of a magnetic field. Using the method of separation of variables, suggested by Schiff and Snyder[7] for a hydrogen-like system in a strong magnetic field, Elliott and Loudon[6] found the eigenfunctions for the orbital motion and the eigenvalues for the diamagnetic exciton energies. The solutions of Eq. (1) were sought, in a cylindrical system of coordinates (ρ, φ, z), in the form of products

$$\Psi(\rho,\ \varphi,\ z)=\frac{e^{im\varphi}}{\sqrt{2\pi}}R_{l,m}(\rho)U^{\nu}_{l,m}(z); \tag{3}$$

here, l is the Landau quantum number, whose values are 0, 1, 2, ...; m is the magnetic quantum number which can have integral values smaller than or equal to l and which is similar to the magnetic quantum number in the quantum-mechanical problem of the motion of a charged particle in a uniform magnetic field.[8] The quantum number ν gives the number and the parity of the excited states of a diamagnetic exciton. We shall consider the values of this number in more detail later

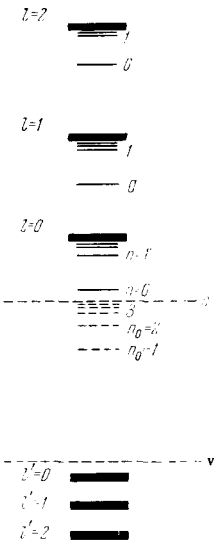

FIG. 2. Schematic representation of the diamagnetic exciton levels in a strong field defined by Eq. (2). The short dashed lines represent a hydrogen-like series of levels of the Wannier–Mott excitons; the long dashed lines represent the conduction and valence band edges in H = 0. When l is increased, the binding energy of the diamagnetic excitons decreases.

in this paper. The first two terms of the product in Eq. (3) are the same functions of ρ and φ as in the absence of the Coulomb coupling. If the field is directed along the z axis, $U^{\nu}_{l,m}(z)$ is a solution of the Schrödinger equation

$$\left[\frac{p_z^2}{2\mu}+V_{l,m}(z)\right]U^{\nu}_{l,m}(z)=\mathscr{E}^{\nu}_{l,m}U^{\nu}_{l,m}(z), \tag{4}$$

where $V_{l,m}(z)$ is an effective potential obtained by averaging the Coulomb potential $-e^2/\kappa r$ over the radial functions. Neither the potential energy $V_{l,m}(z)$ nor the solutions of Eq. (4) can be calculated exactly. Therefore, the problem must be solved making reasonable approximations. Elliott and Loudon[6] approximated the potential $V_{l,m}(z)$ by the following expression:

$$V_{l,m}(z)=-\frac{e^2}{\kappa(a+|z|)}+\frac{Aae^2}{\kappa(a+|z|)^2}, \tag{5}$$

where the quantities a and A depend on the magnetic field and on the quantum numbers l and m. The function $U^{\nu}_{l,m}(z)$ is found to be a solution of a complex differential equation, which can be reduced finally to the Whittaker equation. There are solutions for the bound states, corresponding to the orbital motion of excitons, as well as solutions for free states, corresponding to a continuous spectrum. It is found that transitions to the lowest bound state (the ground state of a diamagnetic exciton) have the strongest intensity and, consequently, they should make the main contribution to the formation of the discrete structure observed in the magnetoabsorption spectrum. Transitions between the Landau subband edges give rise only to a relatively weak continuous absorption background, as shown in Fig. 3 (curve 4). The same figure, which demonstrates the main results of the Elliott-Loudon theory, shows the interband absorption in the absence of a magnetic field for an electron and a hole which are not bound by Coulomb forces (curve 1). Curve 2 in Fig. 3 shows the nature of the absorption when H = 0 but when the Coulomb interaction is taken into account. Finally, curve 3 shows the interband absorption in the presence of a magnetic field only. A shift of the peak (curve 4), corresponding to a transi-

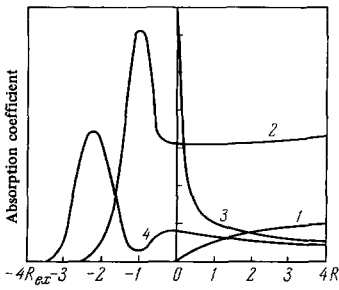

FIG. 3. Comparison of the theoretically predicted absorption edge profile for interband transitions in various cases. The results are taken from [6]. 1) H = 0 and Coulomb interaction ignored; 2) H = 0 and Coulomb interaction allowed for; 3) $H \neq 0$ and Coulomb interaction ignored; 4) $H \neq 0$ and Coulomb interaction allowed for. The curves are calculated on the assumption that $\beta = 2$. The abscissa represents the Rydberg constant of an exciton. It is assumed that the width of the exciton lines is of the order of R_{ex}.

tion of a diamagnetic exciton to the ground state in the direction of the red part of the spectrum indicates an increase of the binding energy of such an exciton in a strong magnetic field.

In analyzing the Elliott and Loudon theory, we note that because of the approximations in the solution of Eq. (4), the magnetoabsorption coefficients calculated using these approximations depend on the parameters a and A; they also depend implicitly (as well as explicitly) on the field H and the quantum numbers l and m. This deficiency of the parametric dependence was removed by Hasegawa and Howard,[9] who solved exactly [using the approximation represented by Eq. (2)] the one-dimensional equation for the Coulomb motion (4). They calculated the exciton levels adjoining the lower Landau sub-bands with $l = 0$ and 1.

Zhilich and Monozon[10] solved the problem on the assumption that the hydrogen-like motion, along the z axis, of a Coulomb pair in excited states with quantum numbers $l = 1, 2, 3, \ldots$ (cf. Fig. 2), is adiabatically slow compared with the motion in a plane perpendicular to the magnetic field. Consequently, in order to separate the variables ρ and z, Zhilich and Monozon used the standard adiabatic approximation, similar to that employed in the theory of molecules in the separation of the nuclear and electronic motion. If the orbital motion of hydrogen-like large-radius states is represented by a quasiclassical frequency $\Omega_n = \mu e^4/\hbar^2 n^3 \kappa^2$, and the motion in a plane perpendicular to the field is represented by the cyclotron frequency $\omega_c = eH/\mu c$, the condition of the validity of the approximation employed is of the form

$$\frac{\varkappa^2 \hbar^3 H}{\mu^2 e^3 c} n^3 \gg 1. \qquad (6)$$

Comparing this expression with Eq. (2), we easily find that it extends the range of validity of the solutions of the problem, compared with the range defined by Eq. (2). Zhilich and Monozon obtained also an explicit dependence of the eigenvalues of the energy of a diamagnetic exciton on l, while in [6,9] the energies of exciton states were calculated only for $l = 0$ and $l = 1$.

In a recent paper, Gor'kov and Dzyaloshinskiĭ[11] investigated the energy of excitons in a strong magnetic field as a function of the momentum of translatory motion.

All the theoretical investigations of the energy states of diamagnetic excitons considered so far have dealt with the idealized case of simple nondegenerate bands. This deficiency, which is common to all of them, is important in comparisons with the experimental data, which mostly have been obtained for crystals with a complex valence-band structure.

Concluding this brief review of the state of the theory of diamagnetic excitons, we must mention that the theory predicts the existence of a series of states of these excitons, converging (like the hydrogen-like series of the Wannier–Mott exciton series) at their photodissociation limits, governed by the energy separations between associated Landau sub-bands (cf. Fig. 2). These series of diamagnetic excitons have a number of special properties. Since the one-dimensional Coulomb potential $e/|z|$ is invariant under the $z \to -z$ transformation, the eigenfunctions should be separable into even and odd. These even and odd states are found to be degenerate in pairs in the case $H \to \infty$, when the interaction between an electron and a hole tends to approach more and more closely the one-dimensional Coulomb coupling; the exception to this rule is the ground state, which is described by a function without nodes and is always even.[6,12] Such twofold degeneracy of states in a "one-dimensional Coulomb series" is similar to the degeneracy in respect of the azimuthal quantum number L in the case of the three-dimensional Coulomb interaction. The energies of the Coulomb motion along z, relative to the limits of the continuous spectrum, are of the form

$$\Delta\mathscr{E}_z^{lm\nu} = -\frac{R_{ex}}{n^2}, \qquad n = 0, 1, 2, \ldots \qquad (7)$$

The quantum number ν, which is shown as a superscript of the function $U^{\nu}_{l,m}(z)$, represents (as already mentioned) the number n of a state in the Coulomb series, as well as its parity: $\nu = 2n$ for the even states; $\nu = 2n - 1$ for the odd states. It follows from Eq. (7) that, in the one-dimensional case, the binding energy of the ground state with $n = 0$ is infinite and corresponds to a capture of a particle by a center. In strong but finite magnetic fields the ground-state energy becomes finite and the twofold degeneracy is lifted so that the excited levels of a diamagnetic exciton with $n = 1, 2, 3, \ldots$, are found to be doublets (Fig. 4) and transitions are allowed only to the even components of these doublets.

The energies of states in a diamagnetic exciton series can be conveniently represented in the form

$$\Delta\mathscr{E}_{ex} = -\frac{R_{ex}}{(n+\delta n)^2}, \quad n = 0, 1, 2, \ldots, \qquad (8)$$

where δn is the quantum defect, which depends on the magnetic field and the quantum number l. This quantum defect represents a correction of the energy of a one-dimensional Coulomb pair in the case of finite values of H. The theory predicts an increase of the binding energy of the discrete exciton states with increasing magnetic field. On the other hand, when the quantum number l increases, the depth of the Coulomb potential well and, consequently, the width of the exciton series decrease, as shown in Fig. 2. We should add that the

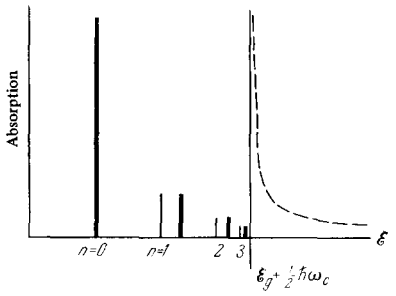

FIG. 4. Schematic representation of a diamagnetic exciton series adjoining the Landau sub-band with $l = 0$. The dashed curve represents the absorption in a magnetic field when the presence of excitons is ignored. The doublet structure of excited states of the diamagnetic exciton with $n \neq 0$ can be seen in the figure. The components of the doublets are designated as in Elliott and Loudon's paper. [6] Thin vertical lines represent the components corresponding to the odd states. They should not appear in the spectrum.

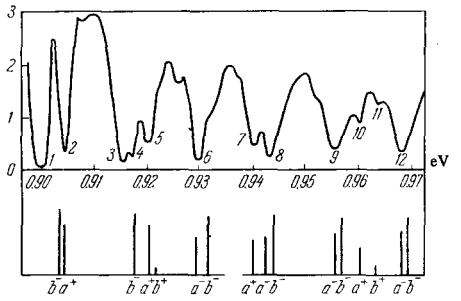

FIG. 5. Part of the magnetoabsorption oscillation spectrum, taken from [14]. The top part of the figure gives the transmission curve, so that the transmission minima (1–12) correspond to the absorption maxima. H = 38.9 kOe, T = 4.2°K. The fourfold [100] axis of a crystal was parallel to a magnetic field and E ∥ H (the π polarization). The lower part of the figure represents a theoretical spectrum [20] and the notation is explained in Fig. 6.

quantum number n is not the same quantum number which represents the states of an exciton in the absence of a magnetic field when the binding energy of states of the Wannier–Mott exciton is given by the expression

$$\Delta\mathscr{E}_{ex} = -\frac{R_{ex}}{n_0^2}, \qquad n_0 = 1, 2, 3, \ldots \tag{9}$$

It is not easy to establish a one-to-one correspondence between the levels of a hydrogen-like series in the absence of a field and in a strong field, as given by the conditions (2) and (6). We can only mention that the states of the diamagnetic excitons with $n \neq 0$ form series which are similar to hydrogen-like series.

Thus, the theory predicts the existence of a diamagnetic exciton series "coupled" to each Landau sub-band. The highest oscillator strength is exhibited by a transition to the lowest state with n = 0 in each of such series. According to Zhilich and Monozon,[10] in the limit of very strong magnetic fields the intensity of transitions to the ground states increases proportionally to H ln H. The intensity of transitions to the excited states with $n \neq 0$ is considerably less and is proportional to $H/\ln^2 H$. This relationship between the intensities of the first and subsequent terms of the series is completely dissimilar from the relationship between the intensities of the terms in the Balmer series of a free exciton, which is obtained in the case of the allowed interband transitions; according to Elliott's calculations,[13] the intensity decreases as n_0^3 when n_0 increases.

We shall now consider the experimental data confirming the conclusion of the exciton nature of the magnetoabsorption oscillation spectra of semiconductors. We shall deal mainly with the experimental data obtained in investigations of the magneto-optical spectra of germanium, particularly the experiments carried out recently by the present authors and A. V. Varfolomeev in the laboratory of E. F. Gross in the A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences.

Germanium satisfies Eq. (2) beginning from fields $H \approx 9$ kOe, because $\kappa = 16$ and $\mu \leq 0.03\, m_0$.

The spectrum of magnetoabsorption oscillations, observed first at T = 4.2°K and investigated in detail by Lax et al.,[14] is shown in Fig. 5. This spectrum has a very complex structure due to the existence of a complex fourfold-degenerate upper valence band at $\mathbf{k} = 0$. The structure of this band in a magnetic field is shown schematically in Fig. 6. A magnetic field lifts completely the fourfold degeneracy and gives rise to four series ("ladders") of the Landau sub-bands. Two of these series are associated with light holes and other two with heavy holes. Details of the structure of the valence band in a magnetic field are known well from numerous investigations of the cyclotron resonance in germanium[15–17] and from theoretical calculations of the valence band of germanium in a magnetic field.[18]

Using these data Burstein et al.,[19] as well as Roth et al.,[20] deduced a theoretical spectrum of the magnetoabsorption oscillations in germanium for transitions between the Landau sub-band edges. The selection rules for the direct parity-allowed interband transitions in germanium are $\Delta l = l - l' = 0, -2$ and $\Delta M = M_S - M_J = 0, \pm 1$.* The second rule determines the nature of the polarization of the spectrum. For obser-

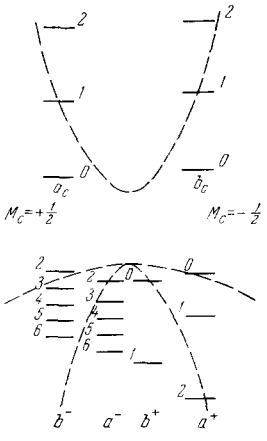

FIG. 6. Schematic representation of the distribution of the diamagnetic Landau sub-bands in the valence and conduction bands of germanium at $\mathbf{k} = 0$. The "ladders" a^- and a^+ of the heavy and light holes have the total momentum projections $M_J = +3/2, -1/2$. The b^- and b^+ "ladders" correspond to $M_J = +1/2, -3/2$.

*) M_S represents the projection of the spin along the field direction and is equal to ±½ for the conduction band; M_J is a quantum number which represents the projection of the total momentum J along the magnetic field direction. For the valence band of germanium, originating from the p states of a free atom, $M_J = \pm 3/2, \pm 1/2$.

vations made at right-angles to the magnetic field, the transitions with $\Delta M = 0$ correspond to the π polarization ($\mathbf{E} \parallel \mathbf{H}$) and the transitions with $\Delta M = \pm 1$ correspond to the σ polarization ($\mathbf{E} \perp \mathbf{H}$).

The experimental spectrum obtained in [14] is compared with the theory in Fig. 5. We can see that in the red part of the spectrum there are strong minima which are not predicted by the theory. It was found subsequently that they were due to the stresses in the thin sample used which split the orbitally degenerate band and distorted the observed effect. These stresses were due to the considerable difference between the thermal expansion coefficients of the transparent substrate and the thin germanium crystal ($d \approx 4\,\mu$) attached to the substrate. Consequently, the germanium sample was deformed at low temperatures.

Figure 7 shows the magnetoabsorption spectrum which we obtained[21,22] for a thin ($d \approx 4$–$6\,\mu$) germanium single crystal free of mechanical stresses. The strong absorption maximum at the longest wavelengths (it is represented by a transmission minimum in Fig. 7) shifted in the direction of short wavelengths proportionally to H^2 (Fig. 8), which was typical of the diamagnetic shift of the Wannier–Mott excitons, because the positions of the Landau levels should obviously be proportional to H. The assignment of the first magnetoabsorption maximum to an exciton state was finally proved by the fact that, when the magnetic field was removed, it still remained in the spectrum and coincided with the well-known absorption maximum corresponding to a direct transition to the 1s state of the Wannier–Mott excitons in germanium.[23]

The theoretical spectrum, calculated in [26], agreed with the experimental data only when the exciton maximum was made to coincide with the first doublet maximum of the theoretical spectrum, corresponding to transitions from the edges of the highest Landau subbands of the b^- "ladder" of the heavy holes and the a^+ "ladder" of the light holes to the electron sub-band with $l = 0$. Then, as is evident from Fig. 7, the theoretical and experimental π spectra exhibited fairly good agreement, at least for the first ten minima. This can be regarded as indicating the common nature of the first and subsequent absorption maxima and, therefore, the dominant role of transitions to exciton states in the interband transitions in a strong magnetic field. The discrepancy between the theory and experiment at higher energies cannot be explained as yet. We may assume that this is the consequence of the effects of the band nonparabolicity, similar to those investigated by Pidgeon and Brown[24] in the magnetoreflection of InSb.

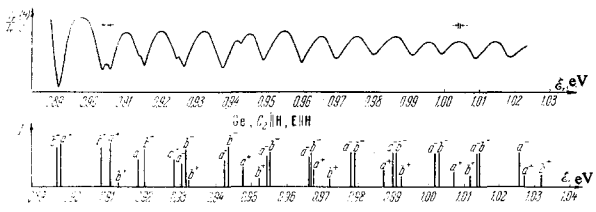

FIG. 7. Experimental and theoretical magnetoabsorption oscillation spectra of stress-free germanium. The top part of the figure shows the transmission curve [22] at T = 4.2°K, obtained in H = 34 kOe, for an n-type sample with an impurity concentration $N = 5 \times 10^{13}\,cm^{-3}$. The theoretical spectrum in the lower part of the figure was calcualted in [19,20] using the band-structure parameters obtained in [17] and the forbidden band width $\mathcal{E}_g = 0.8894$ eV. This value of $\mathcal{E}_g$ was obtained by adding the energy corresponding to the position of the exciton peak at 4.2°K ($\mathcal{E}^0_{ex} = 0.8877$ eV) and the exciton binding energy in H = 0 ($\mathcal{E}^0_{ex} = 0.0017$ eV), calculated theoretically.

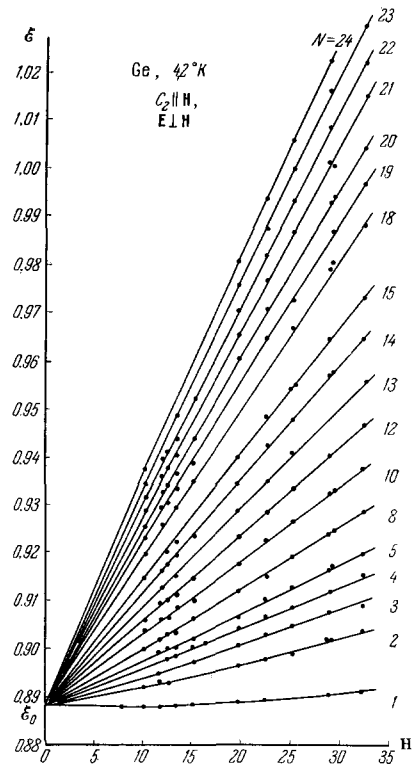

FIG. 8. Dependence of the positions of the magnetoabsorption maxima on the magnetic field intensity (in kOe). N represents serial numbers of maxima in the σ spectrum. The points represent the experimental data. The curves are plotted using $\mathcal{E}_N^{max} = \mathcal{E}_0 + aH^m$, where the parameters a and m are selected by the method of least squares.

For a long time it has been assumed that the magnetoabsorption oscillation effect makes it possible to determine most accurately the forbidden band width (band gap) $\mathcal{E}_g$ of a semiconductor. This is done by plotting a family of straight lines, each of which represents the position of a particular maximum in the magnetoabsorption spectrum as a function of the magnetic field intensity. These lines are extrapolated linearly to a point corresponding to H = 0 (cf. Fig. 8). The value of the energy obtained in this way is assumed to be equal to the forbidden band width. No objections can be raised to this method of determination of $\mathcal{E}_g$ on the basis of the theory associating the magnetoabsorption oscillations with the optical transitions between the Landau sub-band edges. In fact, each of the sub-bands is separated from the edge of the corresponding bands by an amount $\Delta\mathcal{E}_l = (l + {}^1/_2)\hbar eH/m^*c$ in the absence of a magnetic field. Allowance for the spin and the associated additional splitting of the sub-band can only add terms $g\beta_0 H$, which are linear with respect to the field; here, g is the spectroscopic splitting factor and β_0 is the Bohr magneton. Thus, when $H \to 0$, the diamagnetic sub-bands shift toward the band edges linearly with H. However, as

pointed out first by Edwards and Lazazzera,[25] linear extrapolation, to H = 0, of the positions of the maxima for germanium cooled to liquid-nitrogen temperature and free of stresses, gives a value ε_0, which is slightly smaller than ε_g and is close to the position of the exciton absorption maximum in the absence of a field. Moreover, the long-wavelength maxima shift nonlinearly with increasing field intensity.

Figure 8 shows the dependences of the positions of the maxima on the magnetic field obtained in [22] and the curves representing the best approximation of the experimental dependences, obtained by computer analysis. The experimental data are found to be described best by the expression $\varepsilon_N^{max} = \varepsilon_0 + aH^m$, where ε_N^{max} is the energy position of a maximum whose number is N; a and m are functions of the number of the maximum; ε_0 is equal to the energy of the exciton peak ε_{ex}^0 in H = 0. In the range of fields employed in these experiments, the value of m varies, with the serial number of the maximum from m = 2 to m < 1, passing through m = 1. The existence of a linear dependence of the positions of the magnetoabsorption maxima on the magnetic field intensity does not contradict the conclusion about their exciton nature, because at large values of l the binding energy of the diamagnetic excitons decreases, becoming almost independent of H, and the exciton levels shift parallel to the Landau levels in sufficiently strong fields. The existence of values of m smaller than unity may be associated with the band nonparabolicity.

Thus, there are two observations indicating the need to allow for the bound states in an analysis of the magnetoabsorption spectrum in strong fields: 1) the good agreement between the theoretical and experimental spectra is obtained only when the experimentally observed exciton maximum is made to coincide with the theoretical maximum whose wavelength is longest and whose position is calculated on the assumption that there is no Coulomb interaction; 2) it has been established with a high degree of accuracy that the extrapolation of the positions of the magnetoabsorption maxima to H = 0 gives values of $\varepsilon_0 = \varepsilon_{ex}^0$, but not the value of ε_g directly. However, these two observations cannot be regarded as the direct and final solution of the problem.

Experiments on the sudden enhancement of the exciton absorption in doped germanium placed in a magnetic field[26] are more convincing. By doping germanium with impurities, which give rise to sufficiently shallow donor or acceptor levels, we can establish a considerable density of free carriers in the bands at temperatures corresponding to the total or partial ionization of such impurities. Free carriers may screen the Coulomb interaction and in this case the Coulomb potential is replaced by a potential of the type used in the theory of nuclear forces (the Yukawa potential):

$$V(r) = \frac{e^2}{\kappa r} e^{-r/r_d}; \qquad (10)$$

in this expression r_d is the Debye radius, equal to $(\kappa kT/4\pi e^2 N)^{1/2}$, where N is the impurity concentration and k is the Boltzmann constant. When the condition

$$r_d \leqslant a_0^{ex}, \qquad (11)$$

is satisfied (here $a_0^{ex} = \kappa\hbar^2/e^2\mu$ is the Bohr radius of the excitons), the excitons are dissociated because of the screening. When germanium is doped with shallow donors, the condition of Eq. (11) is satisfied by $N_{cr} \approx 10^{16}$ cm^{-3} at T = 77°K, when donors are practically completely ionized. Asnin and Rogachev[27] were the first to observe quenching of the exciton absorption due to the screening of the Coulomb coupling by free carriers in germanium doped with impurities in concentrations close to N_{cr}. Such quenching involved the disappearance not only of the exciton absorption peak but also a change of the absorption edge profile, so that the dependence of the absorption coefficient on the photon energy approached that predicted by Hall, Bardeen and Blatt[28] for noninteracting electrons and holes, when k $\propto (\hbar\omega - \varepsilon_g)^{1/2}$. Thus, doping may "suppress" the Coulomb interaction and reduce the absorption at the edge corresponding to the interband transitions (the curve H = 0 and the dashed line in Fig. 9).

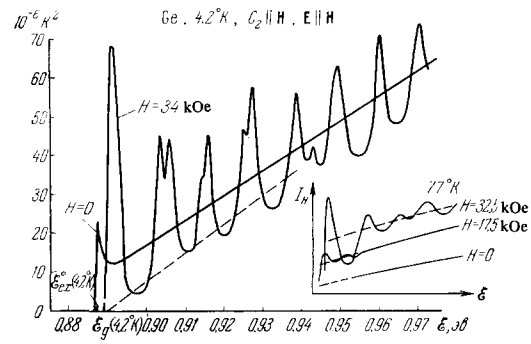

FIG. 9. Dependence of the square of the absorption coefficient of germanium on the energy. The dashed line in the main part of the figure represents the hypothetical form of the spectrum of pure germanium (at 4.2°K in H = 0) in the absence of the Coulomb interaction. The experimentally observed stimulation of the magnetoabsorption in doped Ge, due to an increase in the magnetic field (T = 77° K, N ≈ 8 × 10^{15} cm^{-3}), is shown in the lower right-hand corner of the figure. The dashed line in this case gives the average value of the absorption level.

When the impurity concentration in a germanium crystal is close to N_{cr}, so that the exciton absorption is mostly suppressed, and this crystal is subjected to a magnetic field, a discrete structure is observed at the absorption edge. When the magnetic field is increased, the intensities of these discrete maxima increase, as shown in Fig. 9. Figure 9 demonstrates an important point that, when H is increased, a general rise of the average absorption level (absorption enhancement) takes place. The absorption level tends to the values observed in pure germanium. However, according to the theory of the magnetoabsorption oscillation effect, which ignores the Coulomb interaction, this phenomenon should not be observed. The application of the magnetic field does not alter the average density of quasicontinuous levels in the bands but alters only the detailed distribution of these levels. In such a case, integration of the absorption coefficient as a function of l for H ≠ 0 should yield the same form of the absorption spectrum and the same absorption values as in H = 0, i.e., oscillations of the absorption should be in the form of equal-area "half-waves" increasing in amplitude with increasing field intensity above a constant absorp-

tion level corresponding to H = 0. The observed enhancement of the absorption can be understood in terms of the predominance of transitions to bound exciton states, which are "stabilized" by the magnetic field. Such "stabilization" consists of the appearance of Coulomb-coupled states, when a magnetic field is applied to a crystal in which these states had been destroyed by the screening, and the phenomenon can be understood quite easily in the qualitative sense. In fact, when the strong-field condition of Eq. (2) is satisfied, the Schrödinger equation for the orbital motion of an electron and a hole becomes, as already demonstrated, one-dimensional. In a one-dimensional potential well, we can have a very weakly bound state.[8] Thus, a Coulomb-coupled pair may exist, in a sufficiently strong magnetic field, irrespective of the degree of screening.

The problem of the re-appearance of exciton states in a magnetic field was considered by D'yakonov, Mitchell, and Éfros[29] who followed the ideas given in [6,9] and solved the problem of an exciton in a strong magnetic field for the case when the interaction was described by a potential of the type given by Eq. (10). The main result of their investigation was the establishment of a dependence of the binding energy of the diamagnetic excitons on the concentration of the screening carriers expressed through the Debye radius r_d. The dimension of the ground-state diamagnetic excitons along the magnetic field direction (the length of excitons) decreases, according to [9], with increasing H and in a strong field it is given by the expression

$$r_0 = a_0^{ex}\left[\ln\left(\frac{a_0^{ex}}{\lambda}\right)\right]^{-1},$$

where $\lambda = (\hbar c/eH)^{1/2}$ is the magnetic length, representing the size of the cyclotron orbit, i.e., the exciton is compressed and the screening condition (11) is not satisfied. The energy of the ground state is independent of r_d right up to $r_d \lesssim r_0$. In the region where the Debye radius is less than the length of the ground-state exciton but much larger than the magnetic length λ, the ground-state energy decreases logarithmically when r_d decreases:

$$\mathscr{E}_{ex}^{H} = \mathscr{E}_{ex}^{0}\left[\ln\left(\frac{r_d}{\lambda}\right)^2\right]^2, \quad \text{or} \quad \mathscr{E}_{ex}^{H} \propto \ln^2\frac{H}{N}; \tag{12}$$

when $r_d \ll \lambda$,

$$\mathscr{E}_{ex}^{H} = 4\mathscr{E}_{ex}^{0}\left[\frac{r_d}{\lambda}\right]^4, \quad \text{or} \quad \mathscr{E}_{ex}^{H} \propto \frac{H^2}{N^2}. \tag{13}$$

Thus, it is evident from Eqs. (12) and (13) that at a free-carrier density N, sufficient to screen completely the Coulomb interaction, the application of a magnetic field increases the binding energy of the excitons and thus causes their re-appearance in a magnetic field. A quantitative comparison of the experiment and theory requires much stronger magnetic fields than those employed in [25].

A very convincing proof of the exciton nature of the maxima in the magnetoabsorption spectra was provided by the discovery of the fine structure of the magnetoabsorption oscillations, which was associated with transitions to the excited states of the diamagnetic excitons. Quite recently such a structure was observed by Johnson[30] in the spectra of InSb and Ge in H = 39 kOe. Figure 10 shows part of a transmission spectrum obtained by Johnson, which includes—in addition to the main transmission minimum—a weaker minimum displaced in the direction of higher energies. Using relatively thick germanium crystals ($d \approx 10$–$15\ \mu$) with acceptor concentrations of 2×10^{12} cm^{-3}, we observed a similar structure[31] at liquid-helium temperature. Apart from the additional minimum (minimum 1′ in Fig. 11a), discovered by Johnson, we found another weak minimum 2′, adjoining the short-wavelength side a strong doublet minimum, next to the main minimum. Bearing in mind the distribution of this additional structure in the spectrum and the low intensities of the minima, we associated this structure with transitions to the excited states of the diamagnetic excitons. Thus, the main transmission minimum 1 in Fig. 11a represents a transition to the ground state of a diamagnetic exciton, associated with the lowest electron Landau sub-band with $l = 0$ and the additional minimum 1′—a transition to an even excited state of the same exciton. The second additional minimum 2′ is evidently associated with a transition to an excited state of a diamagnetic exciton belonging to the electron sub-band with $l = 1$.*

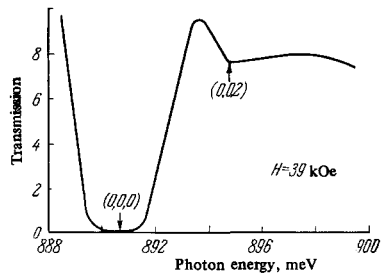

FIG. 10. Transmission curve of a stress-free germanium crystal in H = 39 kOe [30]. The main and additional maxima are designated in accordance with the notation used by Elliott and Loudon.

It is obvious that for reliable detection of the excited states of an exciton, associated with transitions to the higher Landau sub-bands, we need much stronger magnetic fields than those used in our experiments, since the intensities associated with weakly bound states increase with increasing magnetic field.

However, even in relatively weak magnetic fields Johnson observed the structure of the minima at shorter wavelengths, using the electroabsorption technique.[30] A sample free of mechanical stresses was subjected to static (F_1) and alternating ($F_0 \cos \omega t$) electric fields, directed parallel to a magnetic field. The frequency ω of the alternating field was 800 cps. The resultant signal was detected synchronously at the same frequency.

By applying an electric field parallel to the magnetic one, a relatively weak field was expected to ionize those exciton states whose orbits were elongated along a strong magnetic field. It was also natural to expect that the greatest changes in the electric field would be ex-

*)According to Elliott and Loudon, [6] the states of a diamagnetic exciton are represented (as described earlier) by the set of quantum numbers l, m, ν. However, we shall not use this nomenclature of states, which was introduced in order to simplify the band model of a semiconductor.

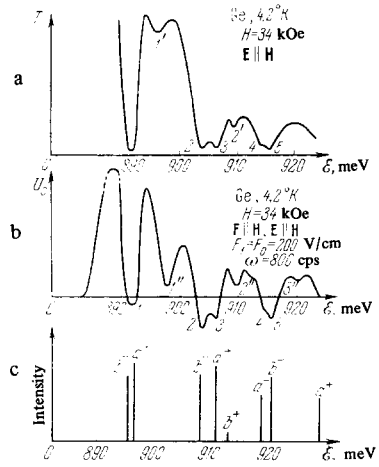

FIG. 11. Structure of the diamagnetic excitons in germanium crystals: [31] a) long-wavelength part of the absorption spectrum, b– electroabsorption spectrum in parallel electric and magnetic fields; c) theoretical magnetoabsorption spectrum of germanium for transmission between Landau subbands, shifted to agree with the main magnetoabsorption maxima.

hibited by the absorption maxima corresponding to the large-radius excited states, as observed by Gross, Zakharchenya, and Kanskaya[32] for the excited states of the Wannier–Mott excitons in Cu_2O. Thus, the excited states of the diamagnetic excitons should be clearly manifested in the differential electroabsorption spectrum. The use of the **F** ∥ **H** geometry avoided also the appearance of new absorption maxima, associated with the stimulation of forbidden transitions in the **F** ⊥ **H** case.[33,34]

Figure 11b shows the electroabsorption curve of germanium in the region where the first five main maxima were observed in the ordinary π magnetoabsorption spectrum. The minima in the differential electroabsorption curve, denoted by 1–5, evidently represented transitions to the ground states of the excitons and coincided with the corresponding minima in the curve of Fig. 11a. Additional minima 1″, 2″, and 3″ appeared clearly. Their positions in the spectra were very close to the energies corresponding to transitions between the Landau sub-band edges, calculated on the assumption that $\mathcal{E}_g$ = 0.8894 eV.

When the magnetic field was increased, the relative intensity of the additional maxima increased. This intensity decreased considerably when the static electric field was increased. At some critical value of the static electric field, the additional minima disappeared altogether, indicating ionization of the corresponding exciton states. The value of the critical field was different for different minima: it lay within the range $F_1 \approx (3.5–5) \times 10^2$ V/m and decreased as the serial number of the minimum decreased. Such a relationship confirmed that the additional minima were associated with the weakly bound excited states of the diamagnetic excitons, which were found to be very sensitive to the applications of an external electric field.

As demonstrated in [35], an electric field applied parallel to a magnetic one, should stimulate the appearance of forbidden transitions to the odd excited states. Indications of the stimulation of such transitions were observed in our experiments.

Thus, careful spectroscopic investigations of the magnetoabsorption spectra of stress-free crystals show that the discrete structure, which appears in a magnetic field, represents transitions to the ground (strong maxima) and excited (additional weak maxima) states of the diamagnetic excitons. We may add that the structure of the spectra should be even more complex because of the spin splitting. The polarization and number of the Zeeman components of the first exciton maximum can be predicted using the ideas of McLean and Loudon.[36] Magnetic fields of about 100 kOe intensity would be required to resolve such a structure.

In conclusion, we shall return again to the experiments concerning the effect of an electric field, parallel to a magnetic one, on the magneto-optical spectrum. We shall now consider stronger static electric fields which affect considerably the strong absorption peaks.[37]

An electric field $F \approx 10^3$ V/cm produced strong "quenching" of the discrete spectrum because of the broadening of the absorption bands and a reduction of the absorption intensity at the main maxima. Oscillations disappeared altogether in fields of about 2.5×10^3 V/cm (Fig. 12). Such "quenching" may be due to the broadening of the Landau sub-band edges due to their shift in the applied electric field (a phenomenon of the Franz–Keldysh effect type). In this case, the field intensity can be estimated using the relationship $h\theta = \Delta\mathcal{E}_{ll'}$, where $\theta = [(eF)^2/2\mu\hbar]^{1/3}$ is a characteristic frequency which occurs in the theory of the Franz-Keldysh effect,[38] and $\Delta\mathcal{E}_{ll'}$ is the separation between neighboring maxima in the magnetoabsorption spectrum. To satisfy this condition for $\Delta\mathcal{E}_{ll'} \approx 10^{-2}$ eV and H = 34 kOe, we require fields $F_{cr} > 10^4$ V/cm.

On the other hand, "quenching" due to the ionization of the exciton states requires that the following equation be satisfied (cf., for example, [39])

$$\left[\frac{\hbar\theta}{\Delta\mathcal{E}_{ex}}\right]^{1/2} = \frac{eFa_{ex}}{\hbar\theta} = 1, \qquad (14)$$

where $\Delta\mathcal{E}_{ex}$ is the binding energy of an exciton. It fol-

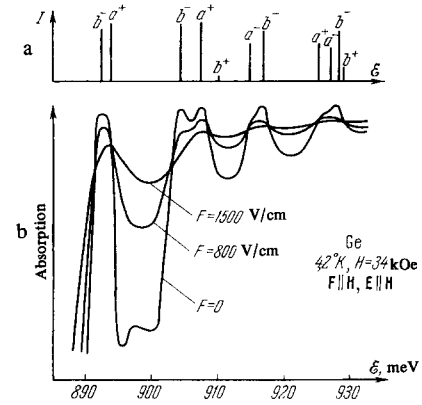

FIG. 12. Influence of an electric field, parallel to a magnetic one, on the spectrum of the magnetoabsorption oscillations of direct transitions in germanium: [37] a) theoretical spectrum of the transitions between the Landau sub-bands of the valence and conduction bands, shifted to fit the experimentally observed maxima; b) experimental spectra at various electric field intensities.

lows from the experimental data that $\Delta\mathcal{E}_{ex}$, which is a function of the magnetic field H and the quantum number l, has a maximum value close to 4×10^{-3} eV. It follows from Eq. (14) that the field necessary for the ionization of the diamagnetic excitons is $F_{cr} \leq 2.8 \times 10^3$ V/cm, which is close to the experimental value.

It is worth noting the apparently strange behavior of the second doublet of the absorption maximum in an electric field. It follows from Fig. 12 that, when the field is increased, the long-wavelength component of the doublet broadens, decreases in intensity, and disappears altogether at a value of the field lower than that at which the short-wavelength component is "quenched." The higher sensitivity of the long-wavelength component to an electric field is observed also in the differential electroabsorption curve (cf. Fig. 11), where the corresponding transmission minimum 2 is deeper. This effect is paradoxical since we established that the long-wavelength component of the doublet is associated with transitions from the Landau sub-band of the heavy holes to the electron sub-band with $l = 1$, and that the short-wavelength component is due to transitions from the light-hole sub-band to the same electron sub-band. The binding energies of the corresponding exciton states should, according to Eq. (8), be proportional to μ, which is the reduced carrier mass, and, consequently, the binding energy should be greater for the heavy holes. Thus, we should expect, theoretically, to observe a "quenching" sequence opposite to that found experimentally.

The contradiction is resolved by using the theoretical results of Wallis and Bowlden,[40] who predicted the existence, in one-dimensional light- and heavy-hole sub-bands, of characteristic quantum effects associated with the degeneracy of the valence band of germanium. According to their calculations, the structure of the diamagnetic sub-bands can be described by the expression

$$\mathcal{E}(l, \zeta) = \mathcal{E}(l) + \frac{\hbar^2 s^2}{2m^*}\zeta^2, \qquad (15)$$

where $\zeta = k_z/s^{1/2}$ is a dimensionless wave number; k_z is the projection of the wave vector onto the direction of the magnetic field; $s = \sqrt{eH/\hbar c} = \lambda^{-1}$ is the reciprocal of the magnetic length. The effective mass m* of each sub-band is given by the curvature of the sub-band and is a function of l and ζ. Figure 13 shows two out of four "ladders" of the Landau sub-bands for the light and heavy holes as a function of ζ (according to [40]). The quantum effects of Wallis and Bowlden for the light-hole sub-band consist of an increase in the values of the effective masses at the point $\zeta = 0$ for low values of l', compared with the value for the mass of $0.04\,m_0$, known from cyclotron resonance experiments. Thus, for the a^+ "ladder," we have $m^*_{0,lt} = 0.12\,m_0$, $m^*_{1,lt} = 0.076\,m_0$, where the numberical subscripts represent l'. As l' increases, m^*_{lt} tends to the value $0.04\,m_0$. In the heavy-hole sub-bands, the quantum effects are particularly dramatic. For the b^- "ladder," we have $m^*_{2,hv} = 0.065\,m_0$ and the effective mass is $m_{s,hv} = 0.023\,m_0$ for $\zeta = 0$, without exhibiting a tendency to approach the value $0.3\,m_0$, known from the cyclotron resonance data. In the a^- series (the notation is explained in Fig. 6), the effective mass for $\zeta = 0$ is even found to have a negative value. Such behavior of the heavy holes in a magnetic field is due, as demonstrated in [40], to the existence of accidental degeneracy in the sub-bands of the a^- and b^- "ladders." In fact, we can see from Fig. 13 that the $a^-(l')$ and $b^-(l'+1)$ sub-bands are very similar from $l = 3$ onwards and coincide at higher numbers.

Figure 13 shows the π transitions between the Landau sub-band edges, which give rise to two different diamagnetic excitons, whose ground states appear in the spectrum in the form of the doublet considered earlier. We can see that, at the point $\zeta = 0$, the heavy-hole sub-band has a stronger curvature than the light-hole sub-band. Consequently, the reduced mass and binding energy of the diamagnetic exciton consisting of a heavy hole and an electron are smaller than, respectively, the mass and energy of the exciton consisting of a light hole and an electron. Using Eq. (15), we can easily show that the ratio of the fields in which the ionization of these excitons takes place is proportional to the ratio of the squares of their reduced masses:

$$\frac{F^{lt}_{cr}}{F^{hv}_{cr}} = \frac{\mu^2_{lt}}{\mu^2_{hv}}. \qquad (16)$$

In our case $\zeta = 10^{-2}$, since $\zeta = k_z/4 \times 10^3\sqrt{H}$. Therefore, in the calculation of the reduced masses of the diamagnetic excitons we should use the values of the effective masses of holes calculated in [40] for $\zeta = 0$. Using the values $m^*_{1,lt} = 0.076\,m_0$, $m^*_{3,hv} = 0.041\,m_0$ and the electron mass $m^*_c = 0.04\,m_0$, we obtain $F^{lt}_{cr}/F^{hv}_{cr} = 2.2$, which is in agreement with the ratio of the ionization fields (1.9) obtained experimentally.

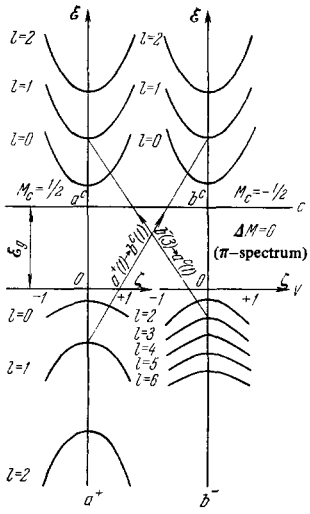

FIG. 13. Schematic representation of the Landau sub-bands in germanium and of the optical transitions between them, which result in the appearance of a doublet structure in the second maximum of the magnetoabsorption spectrum. The sub-band structure is represented in accordance with [40]. Different energy scales are used for the valence and conduction bands.

It must be pointed out that the higher sensitivity, to electric fields, of the exciton maxima corresponding to the heavy holes is observed also in other mangentoabsorption maxima and that it appears in the π as well as in the σ polarization.

Thus, we have considered experiments which prove the exciton nature of the magnetoabsorption oscillations in germanium. Naturally, in strong fields the effects, indicating the existence of the diamagnetic excitons, should be observed also in other semiconductors. The value of the strong field, given by Eq. (2), depends on the properties of a semiconductor. For example, in the case of InSb, the field becomes strong from 2 kOe. As

reported, a fine structure, corresponding to the excited states of the diamagnetic excitons, was observed in this case by Johnson.

The magnetoabsorption oscillation effect has been observed in some semiconducting compounds of the $A^{III}B^{V}$ group (InAs, GaSb, GaAs), but detailed spectroscopic investigations, similar to those described in the present paper, have not been carried out.

It would be of considerable interest to investigate the strong-field magneto-optical effects in CdS and CdSe crystals, which exhibit hydrogen-like series of narrow lines in the absence of a magnetic field. Such investigations would probably make it possible to follow the gradual transition from the hydrogen-like series of a free exciton to the diamagnetic exciton series when the field is increased. However, to observe the structure of the diamagnetic excitons, strong magnetic fields of 10^5 Oe or more would be required for these two compounds. Investigations of cuprous oxide would be particularly interesting because this compound exhibits up to eight terms of a hydrogen-like exciton series in H = 0. In the case of cuprous oxide, the condition (2) is satisfied by fields $H \geq 10^6$ Oe. However, it is possible that the condition (6), similar to (2), may reduce the required values of the strong magnetic fields. It is difficult to obtain an accurate estimate from Eq. (6) because the relationship between the quantum numbers n_0 and n, representing the state of an exciton in the absence of a field and in a strong field, has not yet been definitely established.

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