Diamagnetism of excitons and excitonic molecules in silicon

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(Submitted 30 March 1979)
Zh. Eksp. Teor. Fiz. 77, 752–759 (August 1979)

The emission spectra of excitons and excitonic molecules are studied in silicon crystals strained uniaxially along crystallographic direction ⟨100⟩ in a magnetic field. From the diamagnetic shift in the emission spectrum, the susceptibility of the excitons is estimated. The g factors of the electron and hole in an exciton are determined. It is shown that the diamagnetic susceptibility of an excitonic molecule is close to that of two excitons.

PACS numbers: 71.35. + z, 75.30.Cr, 75.20.Ck

§1. INTRODUCTION

An excitonic molecule (EM), or biexciton, in a semiconductor is a bound state of two electrons and two holes with effective masses m_e and m_h. In the limiting case of equal masses $m_e = m_h$, the stability of the biexciton has been demonstrated by a variational method[1,2] (in this limit, the EM is analogous to a positronium molecule). The model of equal effective masses is quite closely approached by an excitonic molecule in silicon crystals strained uniaxially along direction ⟨100⟩, Si(1–2); such an excitonic molecule has been detected recently in radiative recombination spectra.[3-6] We recall that in Si(1–2) the mean masses of the electron and of the hole are respectively $m_e = (m_{e\parallel} m_{e\perp}^2)^{1/3} \approx 0.33 m_0$ and $m_h \approx 0.23 m_0$. It follows from the experimental observations that the binding energy of a biexciton in Si(1–2) is $\Delta_0 \lesssim 1$ meV,[7,8] whereas the excitonic rydberg is $R_x \sim 12$ meV. Despite such a small binding energy, there is a basis for supposing that the EM remains quite compact. For example, the mean interparticle distances $\langle r_{ee}\rangle$, $\langle r_{hh}\rangle$, and $\langle r_{eh}\rangle$ in an EM, calculated by use of the most successful variational wave function for the case of isotropic and equal masses $m_e = m_h$, are of order[9] $3a_x$ (a_x is the Bohr radius of the exciton).

Because its binding energy is so small, a biexciton in Si(1–2) is a convenient model for experimental investigations of molecular properties in a magnetic field. Thus in magnetic fields ~80 kOe, the binding energy of a biexciton in Si(1–2) is comparable with the energy δE^x of diamagnetic shift of a free exciton and with the energy $g_{e,h}\mu H$ of paramagnetic splitting of the spin sublevels of electrons and holes in a biexciton. An analogous situation for the hydrogen molecule can be expected in astronomically large magnetic fields, of order 10^5 and 10^7 kOe respectively. The question arises whether the biexciton is stable under the action of such magnetic fields. *A priori* it seemed that the magnetic susceptibility of an EM with nearly equal masses m_e and m_h, and therefore "looser" than a molecule formed of hydrogen, should be substantially larger than twice the susceptibility of an exciton. Under these conditions it was expected that the stability of the biexciton would decrease in a magnetic field. Furthermore, two identical particles in a biexciton must be in states with different spins. This also leads to a decrease of the binding energy of the EM with respect to disintegration into two free excitons, in which the electrons and holes are in the spin ground states. From such considerations it followed that even in fields ~50 kOe, excitonic molecules in Si(1–2) would cease to be stable ($\Delta_M \to 0$), and radiation from biexcitons would disappear from the spectra. But experimental observations have shown (§4) that the biexciton radiation band is present in the spectra over the whole range of fields investigated, up to 80 kOe; the relative intensities of the exciton and biexciton radiation lines change little.[1)]

In order to explain this result, which in our view is nontrivial, the present paper investigates in detail the properties of a gas of excitons and biexcitons in a magnetic field. It was found that in Si(1–2), equilibrium between excitons in different spin states is absent in an exciton gas. The g factors of the electrons and holes in excitons were determined, and also the values of the diamagnetic shifts in excitons; and a comparison of measured values with the calculation (§3) was made. Because of the large spin-relaxation time of the electrons and holes in excitons and biexcitons, as compared with the recombination time, it was found that for the binding energy of biexcitons in a magnetic field only the diamagnetic term if important (§3). According to the experiment, it is close to twice the excitonic term (§4).

§2. EXPERIMENTAL METHOD

In the research we investigated pure silicon, with a concentration of residual impurities less than 10^{13} cm^{-3}. To increase the density of the gas phase, we applied uniaxial compression of the Si crystals along the ⟨100⟩ axis.[3] The accuracy of the crystallographic orientation was no worse than 1°. The direction of the magnetic field was either parallel to the axis of compression of the specimens ($\mathbf{H} \parallel \mathbf{P} \parallel \langle 100\rangle$) or perpendicular to it ($\mathbf{P} \perp \mathbf{H} \parallel \langle 100\rangle$). The accuracy of orientation of the specimens along the direction of the magnetic field was about 3°. The research was carried out in fields H up to 80 kOe; the specimen was in superfluid helium at 1.8 K. The equipment for uniaxial compression of the crystals was described in a previous paper.[4]

Nonequilibrium carriers were produced by means of a continuous-action argon laser of power $\lesssim 1$ W. The radiation was recorded with a photomultiplier with cathode S-1, cooled to temperature −60 °C, for which

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the photon-counting mode was insured. The spectral apparatus used was a fast double monochromator with dispersion 10 Å/mm in the working range.

§3. EXCITONS IN A MAGNETIC FIELD

In silicon, indirect radiative disintegration of a biexciton produces a photon, a phonon, and a free exciton.[3] Since in this reaction the exciton state is finite, we shall study primarily the spectroscopic properties of excitons in a magnetic field.

In Si, uniaxial strain along the ⟨100⟩ direction removes the degeneracy in the valence band and in the conduction band as follows. The quadruply degenerate valence band is split into two Kramers doublets with $j=3/2$ and $j=1/2$. The ground state is that with $j=1/2$ and with effective masses $m_{h\parallel}=0.20\ m_0$ and $m_{h\perp}=0.25\ m_0$.[8] In the conduction band, the lowest of six valleys are two located on the compression axis ⟨100⟩, and the remaining four (on the perpendicular axes ⟨010⟩ and ⟨001⟩) are split off by an amount ΔE_c. The effective masses of the electrons remain as before ($m_{e\perp}=0.19\ m_0$ and $m_{e\parallel}=0.95\ m_0$). All four exciton levels are observed in absorption in Si(1–2).[9]

In strained silicon, one can observe luminescence both of "cold" excitons and of "hot," when an electron bound in a hot exciton is in a higher-placed split-off band.[10,11] Accordingly, in the emission spectra at helium temperatures one sees either a single line, if $\Delta E_c > \hbar\omega_{TA}$, or two, if $\Delta E_c < \hbar\omega_{TA}$, where $\hbar\omega_{TA}$ is the energy of an intervalley transverse acoustic phonon. This is due to the fact that the relaxation time of holes from the split-off band to the ground band is much smaller than the lifetime τ_x of an exciton, and the relaxation time of electrons from the higher valleys is large ($<\tau_x$) as long as $\Delta E_c < \hbar\omega_{TA}$.

Emission spectra of cold and of hot excitons in a magnetic field $\mathbf{H}\parallel\mathbf{P}\parallel\langle 100\rangle$ are shown in Figs. 1 and 2 for different states of polarization ($\mathbf{H}\perp\mathbf{E}, \mathbf{H}\parallel\mathbf{E}$). We shall be interested in the relative shifts, in a magnetic field, of the lowest of the split-off spin states. For this purpose, we measured the spectral shift of the red limit of the spectra shown in Fig. 1, as a function of H. The red limit was calculated from the half height. Figure 3 shows the shifts of the emission lines

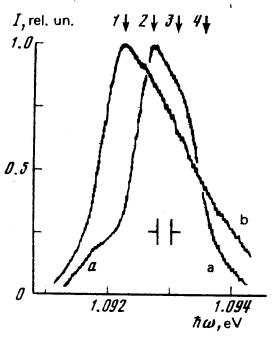

FIG. 1. Recombination spectrum of a cold exciton in magnetic field 80 kOe, $\mathbf{H}\parallel\mathbf{P}\parallel\langle 100\rangle$: a, polarization $\mathbf{E}\perp\mathbf{H}$; b, polarization $\mathbf{E}\parallel\mathbf{H}$. The arrows show the position of the various Zeeman components.

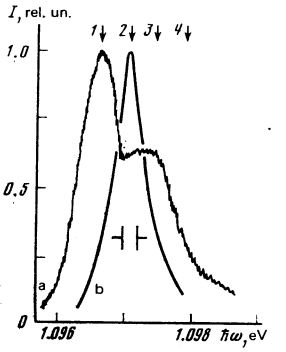

FIG. 2. Recombination spectrum of a hot exciton: a, in magnetic field 80 kOe, $\mathbf{H}\perp\mathbf{P}\langle 100\rangle$; b, without magnetic field.

of excitons in Si(1–2) as functions of the magnetic field for $\mathbf{H}\parallel\mathbf{P}$ and for $\mathbf{H}\perp\mathbf{P}$ and the approximation to these relations by the expression

$$\hbar\omega_x(H)-\hbar\omega_x(0)=\alpha H+\beta H^2. \qquad (1)$$

As is seen from the figure, in fields $H\sim 80$ kOe the terms linear and quadratic in the field are of the same order. The coefficients α and β for the various excitons are given in Table I.

a. Zeeman splitting. The terms linear in the magnetic field, for an exciton in the ground state, can be represented in the form[8]

$$g_e s_z \mu H + g_h j_z \mu H,$$

where $s_z(j_z)$ are the projections of the spin of the electron (hole) and where $g_{e(h)}$ are their g factors. In Si, the g factor of the electron is isotropic and equal to two. At small strains in Si(1–2), g_h can be expressed in terms of g_1 and g_2, the isotropic and anisotropic parts of the g factor in unstrained Si: for $\mathbf{H}\parallel\mathbf{P}$,

$$g_h=g_{h\parallel}=g_1+{}^1/_2 g_2,$$

for $\mathbf{H}\perp\mathbf{P}$,[8]

$$g_h=g_{h\perp}=2g_1+5g_2.$$

In Si, both for free holes and for holes bound to acceptors, $g_2 \ll g_1$.[8,12]

From the values of α_i for the ground state, which are $-\frac{1}{2}(g_e+|g_h|)$, we find on setting $g_e=2$ that in Si(1–2), $|g_{h\parallel}|=1.2\pm0.2$ and $|g_{h\perp}|=2.8\pm0.4$. The value

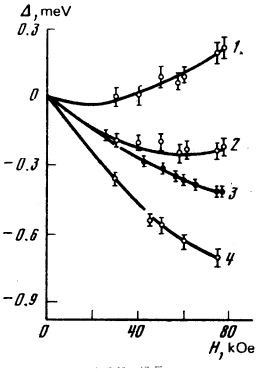

FIG. 3. Variation with magnetic field of the shift of the red limit of the exciton line. 1, line of a cold exciton in polarization $\mathbf{E}\perp\mathbf{H}$ with $\mathbf{H}\parallel\mathbf{P}$; 2, line of a cold exciton in polarization $\mathbf{E}\parallel\mathbf{H}$ with $\mathbf{H}\parallel\mathbf{P}$; 3, line of a hot exciton with orientation $\mathbf{H}\parallel\mathbf{P}\parallel\langle 100\rangle$; 4, line of a hot exciton with $\mathbf{H}\perp\mathbf{P}\parallel\langle 100\rangle$.

TABLE I.

$\alpha\cdot10^2$, meV/kOe	$\beta\cdot10^4$, meV/kOe^2	$\beta_{calc}\cdot10^4$, meV/kOe^2
	Cold excitons H‖P	
0.8±0.2	0.9±0.25	1.18
	Hot excitons H‖P	
0.85±0.15	0.4±0.15	0.45
	H⊥P	
1.3±0.2	0.65±0.25	0.93

$|g_{h\parallel}| = 1.2 \pm 0.2$ is also obtained from the value of the splitting of cold excitons in Si(1–2), which is $g_{h\parallel}\mu H$. If we recover the values of g_1 and g_2 from $g_{h\parallel}$ and $g_{h\perp}$, it is seen that even in excitons $g_1 \ll g_2$; the value $g_1 \approx 1.1$ is close to the value of g_1 for holes bound to B ($g_1 = 1.03$[12]).

b. Diamagnetic shift. It is evident from Fig. 2 that the diamagnetic shift δ^{ex} is anisotropic. This is due primarily to the strong anisotropy of the effective mass of the electron. (As was pointed out above, in Si(1–2) $m_{h\parallel} \approx 0.8\, m_{h\perp}$, whereas $m_{e\parallel} \approx 5\, m_{e\perp}$.) Neglecting the anisotropy of m_h, we have for δ, in the first order of perturbation theory,[13]

$$\delta E^x = \lambda_1 H^2 + \lambda_2 (3H_z^2 - H^2), \tag{2}$$

where

$$\lambda_1 = {}^2/_3 \lambda_0 [(a_\perp^2 + a_\parallel^2) \times (\gamma_h + \gamma_{e\perp}) + a_\parallel^2 (\gamma_h + \gamma_{e\parallel})],$$
$$\lambda_2 = {}^1/_3 \lambda_0 [a_\perp^2 (\gamma_{e\perp} - \gamma_{e\parallel}) + (a_\perp^2 - a_\parallel^2)(\gamma_h + \gamma_{e\perp})], \tag{3}$$

$\lambda_0 = \mu(\gamma_h + \gamma_{e\perp})/4R_x$, $\gamma_i = m_0/m_i$ are the inverse effective masses, and $a_\parallel$ and $a_\perp$ are expressed in units

$$a_0 = \frac{\varepsilon \hbar^2}{e^2 m_0}(\gamma_h + \gamma_{e\perp}),$$

where μ is the Bohr magneton. All the parameters that occur in (3) are known for Si. The calculated values of λ_1 and λ_2 are: $\lambda_1 = 0.68 \cdot 10^{-4}$ meV/kOe^2; $\lambda_2 = 0.25 \cdot 10^{-4}$ meV/kOe^2.

When $\mathbf{H} \parallel \mathbf{P} \parallel \langle 100 \rangle$, for cold and for hot excitons (the electronic ellipsoid is elongated along and transversely to the magnetic field, respectively),

$$\delta E^x_{cold} = (\lambda_1 + 2\lambda_2) H^2,$$

$$\delta E^x_{hot} = (\lambda_1 - \lambda_2) H^2.$$

For hot excitons in Si(1–2) with $\mathbf{P} \perp \mathbf{H} \parallel \langle 100 \rangle$,

$$\delta E^x \approx (\lambda_1 + \lambda_2) H^2.$$

The calculated coefficients for all three cases are given, along with the experimental, in Table I. It is seen from the table that the measured anisotropy for the diamagnetic shift is somewhat smaller than the calculated, but the deviations between the other calculated and experimental values lie practically within the limits of experimental error. If we use the diamagnetic-susceptibility formula

$$\chi = H^{-1} \partial E / \partial H,$$

we get from the experimental data the values $\chi_{cold} = 2.8 \cdot 10^{-25}$ erg/Oe^2 for a cold exciton and $\chi_{hot} = 1.3 \cdot 10^{-25}$ erg/Oe^2 for a hot exciton. For comparison we recall that the diamagnetic susceptibility of a hydrogen atom is $\chi_H = 1.3 \cdot 10^{-30}$ erg/Oe^2.

c. Spin relaxation of excitons. In a magnetic field the degeneracy with respect to spins is removed, and as a result the excitonic ground state in Si(1–2) is split into four (Fig. 3). Under conditions of thermodynamic equilibrium, the intensity of radiation of excitons in excited spin states should decrease, in relaxation to the radiation of excitons in the ground state, as $\exp(\Delta_i/kT)$, where Δ_i is the energy gap between the ground and excited spin sublevels. But it follows from experiment that even at fields $H \sim 80$ kOe, when the distance to the nearest excited spin sublevel amounts to about $3kT$, the intensity of radiation of excitons in the different spin states is of a single order of magnitude. Hence it follows that the spin-relaxation time τ_s in excitons in strained Si is comparable with the lifetime of the excitons ($\sim 10^{-6}$ sec); this is considerably larger than in unstrained Si, where $\tau_s \sim 10^{-8}$ sec.[14] An appreciable increase of τ_s was also observed by us earlier in exciton-impurity complexes during uniaxial compression of Si.[15] It is due to the removal of the degeneracy of the valence band.[16] Allowance for the long spin-relaxation times in Si(1–2) and for the absence of thermodynamic equilibrium between excitons in different spin states is important for understanding of the problem of biexcitons in a magnetic field.

§4. A BIEXCITON IN A MAGNETIC FIELD[17]

If the spins of the electron and of the hole in an exciton can be different, then in a biexciton in semiconductors with nondegenerate bands, two electrons and two holes, in consequence of the Pauli principle, form spin singlets. Accordingly, the energy of an exciton with electron spin s_z and hole spin j_z in a magnetic field is

$$E^x_{s_z, j_z}(H) = E^x(0) + (g_e s_z + g_h j_z)\mu H + \delta E^x, \tag{4}$$

and the energy of a biexciton is

$$E^M_{0,0}(H) = E^M(0) + \delta E^M, \tag{5}$$

where δE^M is the diamagnetic shift ($\sim H^2$) for a biexciton. In Si(1–2), because of the presence of two equivalent valleys in the conduction band, formation of biexcitons with total spin either 0 or 1 is possible (see Fig. 4); therefore in a magnetic field, the ground state is split into a triplet

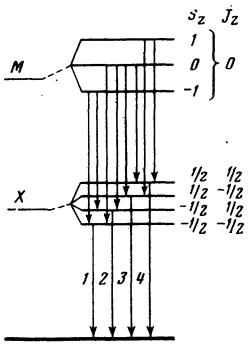

FIG. 4. Schematic diagram of allowed transitions in a magnetic field, illustrating radiative recombination of an exciton (X) and a biexciton (M).

$$E^{M}_{s_z,0}(H)=E^{M}(0)+{}^1/_2 g_e S_z \mu H+\delta E^{M}, \quad (6)$$

where

$$S_z=s_{z1}+s_{z2}=0, \ \pm 1.$$

The binding energy of a biexciton in a magnetic field H is

$$\Delta_H=2E^{x}_{-1/2,\,-1/2}-E^{M}_{-1,0}=\Delta_0-g_h\mu H+2\delta E^{x}-\delta E^{M}. \quad (7)$$

But as was mentioned above, in Si(1–2) the spin-relaxation times are comparable with the lifetime of the excitons. Therefore even when $g\mu H\sim 3kT$, the densities of excitons in the different spin states are of the same order of magnitude. In this case, in Si(1–2) in the presence of a magnetic field those states of the biexcitons are advantageous in which the holes form a spin singlet. Then the binding energy of a biexciton is determined by the expression

$$\Delta_H=-(E^{M}_{s_{z1}+s_{z2},0}-E^{x}_{s_{z1},1/2}-E^{x}_{s_{z2},\,-1/2})=\Delta_0+2\delta E^{x}-\delta E^{M}. \quad (8)$$

The emission spectrum of a gas of excitons and biexcitons in a magnetic field $H=60$ kOe is shown in Fig. 5. The emission lines are greatly broadened, but the structure of the individual Zeeman components is not resolved, and the value of Δ_H cannot be determined from the spectrum. It can only be stated that $\Delta_H>0$. The change of the binding energy of a biexciton in a magnetic field can also be estimated from the change of the ratio of total emission intensities of biexcitons and excitons, I_M/I_x. Under conditions of thermodynamic equilibrium between excitons and biexcitons, $I_M/I_x\sim n_M/n_x\sim e^{\Delta/kT}$. The ratio I_M/I_x does not change by more than a factor 3 over the whole measurement range $H\lesssim$ kOe, either for $\mathbf{H}\parallel\mathbf{P}\parallel\langle 100\rangle$ or for $\mathbf{H}\perp\mathbf{P}\parallel\langle 100\rangle$. Since the mean rate of excitation varies only slightly with magnetic field for fixed pumping, $|\Delta_0-\Delta_H|$ does not exceed $(2/3)kT$, or ~0.4 meV. According to (8),

$$|\Delta_0-\Delta_H|=|2\delta E^{x}-\delta E^{M}|.$$

It was found above that for cold excitons, $\delta E^{x}(80\text{ kOe})=0.7$ meV for $\mathbf{H}\parallel\mathbf{P}$ and 0.4 meV for $\mathbf{H}\perp\mathbf{P}$. Thus the biexcitonic diamagnetic shift does not exceed three times the excitonic. The experimentally established upper bound to the ratio of the susceptibilities of the biexciton and of the exciton is a direct indication that the electron-hole correlations in the biexciton, which

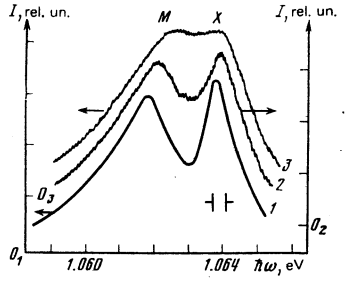

FIG. 5. Spectrum of recombination radiation of an exciton and of a biexciton: 1, without magnetic field; 2, in magnetic field 80 kOe and polarization $\mathbf{E}\perp\mathbf{H}$; 3, in polarization $\mathbf{E}\parallel\mathbf{H}$ at $H=80$ kOe.

is analogous to a positronium molecule, are extremely strong.

Such a small value of δE^{M} for a weakly bound state seems at first glance unexpected. But one must pay attention to the fact that the diamagnetic shift for an excitonic molecule is not determined solely by the Langevin term, calculated in the first order of perturbation theory,

$$\delta E_L^{M}=\frac{e^2}{8c^2}\sum_i \frac{\langle 0|[\mathbf{H}\mathbf{r}_i]^2|0\rangle}{m_i}, \quad (9)$$

where i is the index of the various particles that make up a biexciton, and where $|0\rangle$ is the vector of the ground state of the biexciton. Because the wave function of the molecule is not an eigenfunction of the magnetic moment, the term linear in the magnetic field gives a negative contribution to the energy of the ground state of the biexciton in the second order (the Van Vleck term):

$$\delta E_{VV}^{M}=\left(\frac{1}{2c}\right)^2\sum_n \frac{\left|\langle n|\sum_i \mathbf{H}[\mathbf{r}_i\mathbf{j}_i]|0\rangle\right|^2}{E_0-E_n}, \quad (10)$$

where $|n\rangle$ is the vector of the excited state, and where $\mathbf{j}_i=e_i\mathbf{p}_i/m_i$ is the current operator of the ith particle.

Unfortunately, because of the summation over all excited states, a calculation by formula (10) is difficult. Édel'shtein[18] used the fact that with a suitable canonical transformation of the coordinates, one can effect a decrease in absolute value of the term δE_{VV}^{M}. Then formula (9) gives a more accurate upper bound to the diamagnetic shift, since $\delta E_{VV}^{M}<0$. Édel'shtein's calculation,[18] the canonical coordinates used are not the coordinates of all the particles with respect to the center of mass, but the relative coordinates of the centers of mass of the excitons and the relative coordinates of the electron and hole in each exciton. The wave function used as $|0\rangle$ was that of Brinkman, Rice, and Bell[7]; the negative term δE_{VV}^{M} was not taken into account. The value obtained was $\delta E^{M}<3.8\delta E^{x}$, which does not contradict the experimental result $\delta E^{M}<3\delta E^{x}$.

In conclusion, it should be noted that in the limit of a strong field H, when the contribution to the energy from the magnetic field far exceeds the energy of Coulomb interaction, the stability of the biexciton should, according to the calculation of Ref. 19, increase as $\Delta_H\sim\ln^2 H$. The calculation in this limit of magnetic fields relates to the stability of excitonic molecules with resultant spins unity both for the electrons and for the holes; these are completely unstable at $H=0$. The experiments performed in our research do not exclude the possibility that the binding energy of a biexciton in Si(1–2) increases monotonically with increase of magnetic field.

In conclusion, the authors express their thanks to G. E. Pikus and V. M. Édel'shteĭn for fruitful discussions.

[1)]Some of the material contained in §4 was published by us earlier in conference proceedings.[17]

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Translated by W. F. Brown, Jr.

Diamagnetic susceptibility of excitonic molecules

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(Submitted 30 March 1978)
Zh. Eksp. Teor. Fiz. 77, 760–763 (August 1979)

Excitonic molecules in semiconductors in the presence of a magnetic field are considered. In the limit of equal isotropic masses of the electron and hole it is shown that the susceptibility of the molecule can exceed the susceptibility of two free excitons by not more than a factor of 1.9.

PACS numbers: 71.35. + z, 75.20. − g

Generalizing the exciton concept, Lampert[1] postulated the possible existence, in semiconductors, of various complexes consisting of a larger number of particles than the electron-hole pair. One such complex is a bound state of two electrons and two holes—the excitonic molecule (EM). If the effective masses of the electron and hole differ substantially, then the EM is equivalent to the hydrogen molecule. By contrast, the possibility of binding four particles with equal masses is less obvious. This problem, which is equivalent to the problem of the existence of the positronium molecule was finally solved by Hylleraas and Ore[3] by a variational method.

It has recently become possible to investigate EM in experiment. The most convincing proof of observing EM pertains to Si.[3-5] Although the electrons in Si have an isotropic mass, the average electron mass $m_e = (m_{e\perp}^2 m_{e\parallel})^{1/3} \approx 0,33 m_0$ is almost equal to the hole mass $m_h \approx 0,23 m_0$. Therefore the model of equal isotropic masses seems suitable for Si. We consider throughout just this limiting case. The degeneracy of the hole band is assumed lifted by external pressure.

It follows from Refs. 2 and 6, where a more successful trial wave function was proposed, that the molecule has a rather small binding energy $\Delta \approx 0.03 R_x$ where R_x is the exciton binding energy, and relatively large dimensions of order $3a_x$, where $a_x = 2\varepsilon_0 \hbar^2/me^2$ is the Bohr radius of the exciton. Two contradictory assumptions can be made here with respect to the value of the diamagnetic susceptibility χ_M of the molecule. On the one hand, in analogy with atoms, where according to Langevin the diamagnetic susceptibility is proportional to the square of the distance from the electron to the nucleus one might expect $\chi_M \sim 10\chi_x$, where χ_x is the diamagnetic susceptibility of the exciton. On the other hand, the low binding energy seems to indicate that the electron-hole correlations in the molecule are practically the same as in the exciton and therefore χ_M should not differ greatly from $2\chi_x$. The result of the present paper is the inequality

$$|\chi_M/2\chi_x| < 1.9. \tag{1}$$

The Hamiltonian of a molecule in a magnetic field is

$$\mathcal{H} = \frac{1}{2m}\left[\left(\mathbf{p}_1 + \frac{e}{c}\mathbf{A}_1\right)^2 + \left(\mathbf{p}_a - \frac{e}{c}\mathbf{A}_a\right)^2 + \left(\mathbf{p}_2 + \frac{e}{c}\mathbf{A}_2\right)^2 + \left(\mathbf{p}_b - \frac{e}{c}\mathbf{A}_b\right)^2\right] + V_{coul}, \tag{2}$$

where $\mathbf{r}_1$ and $\mathbf{r}_2$ are the coordinates of the electrons, $\mathbf{r}_a$ and $\mathbf{r}_b$ are the coordinates of the holes, $\mathbf{A}_r = 1/2\mathbf{H} \times \mathbf{r}$, and V_{coul} is the Coulomb-interaction potential. We have

 0038-5646/79/080384-03$02.40