# Effect of exciton motion in a magnetic field on luminescence; indirect forbidden transitions in Ge

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(Submitted 6 June 1983)
Zh. Eksp. Teor. Fiz. **86**, 338–342 (January 1984)

The effect of exciton motion in a magnetic field on the luminescence intensity has been studied in germanium compressed approximately along the ⟨100⟩ axis. This effect was studied by comparing the intensity of a forbidden transition involving the emission of a *TA* phonon with the intensity of an allowed *LA* transition. The increase in the ratio $I_{TA}(H)/I_{LA}(H)$ with increasing magnetic field is explained by a magnetic Stark effect. The calculated functional dependence $I_{TA}(H)/I_{LA}(H)$ agrees well with the experimental results at $H \leqslant 3$ T.

With their long lifetimes and large first Bohr radius, indirect excitons in highly compressed germanium crystals with simple bands are extremely attractive for studying the effect of a magnetic field on the properties of such excitations. The basic method for studying an exciton system in an indirect semiconductor is to study the luminescence. The radiative recombination of indirect excitons in germanium is accompanied by the emission of phonons, which carry off the Brillouin quasimomentum. In germanium, such transitions may be either allowed [the corresponding transmission matrix element is $g_a(\mathbf{k}_e, \mathbf{k}_h) \neq 0$ at $\mathbf{k}_e = \mathbf{k}_h = 0$] or forbidden [$g_f(0, 0) = 0$]. The quasimomenta of the electrons and holes are reckoned from the minima of the corresponding energy bands. Among the allowed transitions are those involving the emission of odd *TO* and *LA* phonons, while transitions involving the emission of even *TA* and *LO* phonons are among the forbidden transitions.[1] In previous studies of allowed transitions, the Zeeman splitting and diamagnetic shift of excitons have been measured.[2,3] The change in the annihilation probability in a magnetic field has also been measured.[4]

In the present experiments we arranged a situation in which the free nature of the motion of excitons is expressed most completely. We made use of a particular feature of the band structure of germanium: the existence of both forbidden and allowed transitions, because the minimum of an electron valley lies at a symmetric point at the boundary of a Brillouin zone.

## 2. EXPERIMENTAL PROCEDURE AND RESULTS

We studied pure germanium crystals with a residual impurity concentration of $2\cdot10^{11}$ cm$^{-3}$. The nonequilibrium excitons are excited by a YAG laser. The exciton emission is detected by a cooled Ge(Cu) photoresistance in synchronous-detection operation. The double monochromator has a dispersion of 8Å/mm. The technique for applying a uniform uniaxial compression to the crystals is described in Ref. 5. The magnetic field is directed along the compression axis, $\sim$⟨100⟩.

At liquid-helium temperatures the intensity of the emission of excitons involving the emission of *TA* phonons in germanium is very low. In undeformed germanium crystals, the intensity ratio of the *TA* and *LA* components is 1/100 (Ref. 1). This ratio is reduced by another factor of several units in uniaxially compressed crystals. This circumstance apparently explains why this line has not been studied previously. In order to study the *TA* component experimentally, we would like to raise the exciton density. This approach, however, raises the fraction of excitonic molecules in the exciton gas, and the emission line of these molecules partially overlaps the free-exciton emission line.[2] We accordingly worked at a relatively high temperature, $T \approx 2.15$ K, and we restricted the excitation level to $W \leqslant 2$ W/cm$^2$. Figure 1 shows the emission spectrum of germanium highly compressed along the ⟨1,1,16⟩ axis under these conditions. We see that the intensity of the violet tail due to the emission of excitonic molecules[2] is quite low in the allowed component. As expected for the emission of excitonic molecules, the relative intensity of this tail increases slightly in the forbidden component. The reason is that the emission probability is higher for excitonic molecules than for excitons.[2]

In a weak magnetic field, the binding energy of the excitonic molecules decreases, and they decompose into free excitons.[2] The violet tail observed on the exciton accordingly vanishes at $H > 0.5$ T in both the *LA* and *TA* components. To eliminate from consideration effects of a change in the exciton density in the weak magnetic field we restricted the study to the ratio of the intensities of the forbidden and allowed components.

Figure 2 shows the magnetic-field dependence of the ratio of the integrated exciton emission intensities in the for-

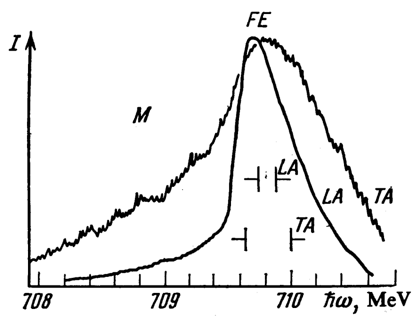


FIG. 1. Allowed (*LA*) and forbidden (*TA*) components of the emission spectrum of pure germanium crystals compressed along the ⟨1,1,16⟩ axis ($p \approx 330$ MPa) at 2.2 K. For clarity the *TA* component has been shifted along the energy scale by 19.8 meV, the difference between the energies of the *TA* and *LA* phonons.


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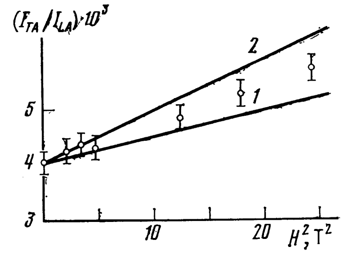


FIG. 2. Comparison of the experimental (points) and theoretical (lines) behavior of the ratio of the integrated intensities of the free-exciton emission in the forbidden and allowed components as a function of the magnetic field. The calculated results are normalized to the experimental value of $I_{TA}/I_{LA}$ at $H=0$. Curve 1 was derived for $|\mathbf{b}| = |\mathbf{c}|$, and curve 2 for $|\mathbf{b}| = |\mathbf{c}|/2$.

bidden and allowed components, $I_{TA}(H)/I_{LA}(H)$. This ratio increases with increasing magnetic field.

## 3. THEORETICAL COMMENTS AND DISCUSSION

The amplitude for the annihilation of an exciton with a momentum **p**, accompanied by the emission of a phonon with a momentum **q** (reckoned from the minimum of the electron valley), can be written in the form

$$A=\sum_{\mathbf{k}_e\mathbf{k}_h}\delta(\mathbf{k}_e+\mathbf{k}_h-\mathbf{q})\,g(\mathbf{k}_e,\mathbf{k}_h)\langle\mathbf{k}_e,\mathbf{k}_h|\mathbf{P}\rangle, \tag{1}$$

where

$$g_a(\mathbf{k}_e,\mathbf{k}_h)\approx g_0, \qquad g_f(\mathbf{k}_e,\mathbf{k}_h)\approx\mathbf{n}_e\mathbf{k}_e+\mathbf{n}_h\mathbf{k}_h, \tag{2}$$

$\langle\mathbf{k}_e,\mathbf{k}_h|\mathbf{P}\rangle$ is the Fourier transform of the exciton wave function in a magnetic field,[6]

$$\langle\mathbf{r}_e,\mathbf{r}_h|\mathbf{P}\rangle\equiv\Psi_P(\mathbf{R},\mathbf{r})=\exp\left\{i\left(\mathbf{P}+\frac{e}{c}\mathbf{A}(\mathbf{r})\right)\mathbf{R}\right\}\Psi_P(\mathbf{r}),$$
$$\mathbf{R}=M^{-1}(m_e\mathbf{r}_e+m_h\mathbf{r}_h),\qquad \mathbf{r}=\mathbf{r}_e-\mathbf{r}_h,\qquad M=m_e+m_h. \tag{3}$$

To simplify the calculations we assume that the dispersion law for the electrons and holes is isotropic. From (1) and (3) we find

$$A_a=g_0\delta(\mathbf{P}-\mathbf{q})\Psi_P(0), \tag{4a}$$

$$A_f=\delta(\mathbf{P}-\mathbf{q})\left[(\mathbf{bP})\Psi_P(0)-i(\mathbf{c}\nabla)\Psi_P(0)\right], \tag{4b}$$

$$\mathbf{b}=(\mathbf{n}_e m_e+\mathbf{n}_h m_h)M^{-1},\qquad \mathbf{c}=\mathbf{n}_e-\mathbf{n}_h. \tag{4c}$$

At low temperatures, all the excitons are in the ground state. In the absence of a magnetic field, the ground-state wave function of an exciton with a center-of-mass momentum **p** is symmetric with respect to an inversion of coordinates, and it remains so when there is an anisotropy in the effective masses. In this case we have $\nabla\Psi_P(0)=0$, and the only contribution to the luminescence in the forbidden component comes from the first term in (4b). The situation changes substantially when a magnetic field is imposed. The Hamiltonian for the function $\Psi_p(r)$ from (3) is[7]

$$\mathscr{H}_P(\mathbf{r})=\frac{\mathbf{P}^2}{2M}+\frac{2e}{MC}\mathbf{PA}(\mathbf{r})+\mathscr{H}_0(\mathbf{r}), \tag{5a}$$

$$\mathscr{H}_0(\mathbf{r})=\frac{1}{2\mu}(-i\nabla)^2+\frac{e}{2\mu c}\gamma\mathbf{H}\hat{\mathbf{l}}+\frac{e^2}{8\mu c^2}[\mathbf{H}\times\mathbf{r}]^2-\frac{e^2}{\varepsilon_0 r}, \tag{5b}$$

$$\mathbf{A}(\mathbf{r})={}^1/_2[\mathbf{Hr}],\qquad \gamma=(m_e-m_h)/(m_e+m_h),\qquad \mu=m_e m_h/M.$$

The operator $\mathscr{H}_0(\mathbf{r})$ describes the diamagnetic properties of an exciton at rest. For $\hbar\omega_c<\mathrm{Ry}$, where $\omega_c=eH/\mu c$ and $\mathrm{Ry}=\mu e^4/2\varepsilon_0^2\hbar^2$, a magnetic field leads to a transverse compression of the ground-state wave function,[4]

$$\Psi_0(\mathbf{r})=\frac{\exp(-r/a)}{\pi^{1/2}a^{3/2}}\left\{1+\left(\frac{\hbar\omega_c}{4\mathrm{Ry}}\right)^2\left[\frac{11}{6}-\frac{r^2}{6a^2}\left(1+\frac{3}{2}\sin^2\vartheta\right)-\frac{r^3}{6a^3}\sin^2\vartheta\right]\right\}, \tag{6}$$

and thus an intensification of the luminescence. Here $\vartheta$ is the angle between the radius vector and the direction of the magnetic field. According to (4a), this is the entire effect of a weak field on exciton optics for allowed transitions. The effect of the second term in (5a),

$$\frac{2e}{Mc}\mathbf{PA}(\mathbf{r})\equiv e\mathbf{Er},\qquad \mathbf{E}=\left[\frac{\mathbf{P}}{Mc}\times\mathbf{H}\right], \tag{7}$$

can also be calculated by perturbation theory if the magnetic field is not too strong. This term causes a so-called magnetic Stark effect[8,9]: the appearance of an electric field which acts on an exciton which is moving in a magnetic field at a velocity $\mathbf{P}/M$. This field polarizes the exciton[1)]: The eigenfunction $\Psi_0(\mathbf{r})$ of the operator $\mathscr{H}_0(\mathbf{r})$ acquires a term

$$-\Psi_0(\mathbf{r})\frac{\varepsilon_0 a}{e}\mathbf{Er}\left(1+\frac{r}{2a}\right),\qquad a=\frac{\hbar^2\varepsilon_0}{\mu e^2}. \tag{8}$$

This correction is found by the method of Refs. 11 and 12; it leads to

$$\nabla\Psi_P(0)=-\Psi_P(0)\varepsilon_0\mathbf{E}a/e. \tag{9}$$

We also find an increment in the kinetic energy,

$$-\frac{9}{4}a^3\varepsilon_0E^2=-\frac{\mathbf{P}_\perp^2}{2M}\left[\frac{9\mu}{8M}\left(\frac{\hbar\omega_c}{\mathrm{Ry}}\right)^2\right], \tag{10}$$

which means that a magnetic field increases the translational mass of the exciton in the directions perpendicular to the field **H**. The derivation of Eq. (8) makes use of the circumstance that, because of the odd parity with respect to inversion, the correction (7) to the wave function does not alter its normalization with an accuracy to terms proportional to $H^2$ inclusively. From (4), (8), and (9) we find an expression for the intensity ratio:

$$I_f(H)/I_a(H)=\langle(\mathbf{bP})^2+(\mathbf{c},\varepsilon_0\mathbf{E}a/e)^2\rangle, \tag{11}$$

where the angle brackets denote an average over a Boltzmann distribution. A magnetic-field dependence arises twice in (11): first, because $E\sim H$; second, because of the redistribution of excitons in momentum space due to a change in the translational mass of the exciton, described by (10).

The forbidden transition thus intensifies more rapidly than the allowed transition with increasing magnetic field, exclusively because of the magnetic Stark effect. In other words, it is a demonstration of the free motion of excitons.

Under our experimental conditions the average thermal energy of the excitons is $(T\approx 2\ \mathrm{K})\ v_T=1.7\cdot 10^6$ cm/s; this value corresponds to an electric field $E\approx 170$ V/cm at $H=1$ T.

Let us compare the theoretical functional dependence $I_f(H)/I_a(H)$ for excitons in germanium with the experimental data (Fig. 2). For this calculation we use the values $\mu/M = 1/8$ and $a = 160$ Å (Ref. 3). For the ratio of parameters we choose the values $|\mathbf{c}|/|\mathbf{b}| = 1$ (curve 1) and $|\mathbf{c}|/|\mathbf{b}| = 2$ (curve 2). The value $|\mathbf{c}|/|\mathbf{b}| \approx 2$ follows from (4c) if we assume $n_e^2/n_h^2 \leqslant 1/10$ in accordance with the experimental data of Refs. 1 and 13.

The weak-field approximation, $\hbar\omega_c <$ Ry, breaks down at $H \sim 1$ T. Accordingly, the separate experimental curves of $I_{TA}(H)$ and $I_{LA}(H)$ in fields above this value can no longer be described by equations in which the effect of H on $\Psi_0$ is taken into account in first-order perturbation theory.[4] In contrast, the ratio $I_{TA}(H)/I_{LA}(H)$, in which the factor $|\Psi_0(0)|^2$ cancels out, can be described well by expression (11) over a broader field range (Fig. 2).

We wish to emphasize that there is a distinction between the magnetic properties of atomic gases and excitons in semiconductors. The corrections $\sim H^2$ to the wave function of an exciton at rest are determined by the reduced mass $\mu$ and are thus analogous to the corresponding corrections in atoms. The corrections stemming from the magnetic Stark effect, in contrast, are determined by the total mass $M$, so that they are important only for excitons; they are negligibly small for ordinary atoms. The only exceptional cases might arise in a study of highly forbidden atomic transitions.

We wish to thank I. V. Kukushkin and V. B. Timofeev for useful discussions.

[1)]The polarization of an exciton due to motion in the limit of strong magnetic fields, $\hbar\omega_c \gg$ Ry, was first mentioned and calculated in Ref. 10.

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Translated by Dave Parsons