Spin-oriented excitons in uniaxially strained germanium in a strong magnetic field

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Luminescence spectra have been measured in an effort to learn about the behavior of indirect excitons in ultrapure Ge crystals strained uniaxially and uniformly along a direction near ⟨100⟩ in magnetic fields up to 14 T. The shift and intensification in a magnetic field of the *LA* line of indirect recombination of excitons have been studied. A new emission line is observed on the low-energy side of the free-exciton line.© *1995 American Institute of Physics.*

Since the binding energy of an exciton is small, one can satisfy the strong-field condition, i.e., the condition that the paramagnetic energy of the electron and hole making up the exciton be greater than the binding energy of the exciton, in fields which are actually attainable. An exciton in a strong field has attracted interest because of a restructuring of the intrinsic state of the exciton and a change in the nature of the interaction between excitons. In the strong-field limit the transverse dimension of an exciton is determined by the magnetic length $l_H = eH/\hbar c$, while the longitudinal dimension changes considerably more slowly $[\sim \ln^{-1}(H/H_c)]$. As a result, an exciton comes to look like a cigar pointed along the field, and the effective volume of the exciton decreases. A strong magnetic field suppresses the van der Waals interaction between excitons, but increases the quadrupole moment of the exciton. This effect could in principle result in a binding of a pair of excitons in a triplet molecule[1,2] and in the formation of chains of such molecules. A possible Bose condensation has been predicted in a gas of triplet molecules.[2]

Uniaxially strained Ge is a convenient model system for studying the properties of free excitons, an electron–hole liquid, and exciton molecules (biexcitons). In the case of a pronounced, uniaxial, uniform deformation ($P > 1$ kbar) along ⟨111⟩ directions and approximately along ⟨100⟩ directions (we denote this case by ⟨∼100⟩), the valley degeneracy in the conduction band is lifted, and degenerate states in the valence band split. The band structure becomes simpler. The hole dispersion relation, which is extremely complex in unstrained Ge, becomes approximately parabolic when there is a sufficiently large level splitting in the valence band. As was shown in Ref. 3, a uniaxial, uniform strain along an ⟨∼100⟩ axis makes it possible to destabilize an electron–hole liquid, because of the more effective decrease in the density of states in the valence band. It becomes possible in this case to produce a gas of excitons with a density up to 5×10^{15} cm^{-3} at $T \sim 2$ K. Attempts to raise the density further may fail because a gas–liquid interface is reached or because the threshold for a Mott transition is reached (the parameter r_s has a value ≈ 2). A magnetic field shifts the threshold for the Mott transition to a

density several times higher, even in weak fields.[4] The stabilization of an electron–hole liquid in a strong magnetic field in uniaxially strained Ge $\langle\sim 100\rangle$, on the other hand, remains an open question.

The magnetic field H_c, found by equating the sum of the energies of the electron and the hole in the zeroth Landau level to the exciton Rydberg Ry, turns out to be 2.9 T for Ge $\langle\sim 100\rangle$. Here we are taking the cyclotron mass of the holes in the limit of an infinite pressure. In a field of 14 T we would have a ratio $H/H_c \cong 5$.

For the present measurements we used germanium single crystals with a concentration of electrically active impurities no greater than 10^{11} cm^{-3}. The test samples were rectangular parallelepipeds with linear dimensions of $2.5\times 2.5\times 10$ mm, cut in such a way that the long direction was parallel to the $\langle 1,1,16\rangle$ axis within 1°. Samples were treated in the polishing etchant SR-4A before being placed in the cryostat in order to reduce surface recombination.

A brass jig consisting of a ring, a die, and a support was used to produce a uniform uniaxial deformation. The die which was inserted into the ring exerted pressure on the test sample which was attached to the support. The length-to-diameter ratio of the die was about 3.5. The surfaces of the die and the ring were polished to an optical finish and carefully matched to each other. A gasket of tin–lead solder (50% Sn + 50% Pb) 1 mm thick was inserted between the crystal and the die, and a similar gasket was inserted between the crystal and the support. The crystal was held in place between the gaskets and was pressed ~ 0.5 mm into them at a low pressure at room temperature. The samples were then immersed directly in liquid helium. Optical fibers were used to bring the exciting light to the crystal and to extract the luminescence signal. A fiber with a quartz core 0.4 mm in diameter turned out to be optimum for the purpose. The fiber was curved slightly and brought up at a certain angle to a mirror attached to the support of the sample in such a way that the reflected light was incident normally on the surface of the crystal. The optical path length between the end of the fiber and the surface of the crystal was less than 0.5 mm. The transmission of the particular fiber which we used was $\sim 70\%$ (reflection from the ends is being taken into account here) at the wavelength 5145 Å, while the transmission in the region of exciton luminescence was no worse than 50%. The mirror reflected 50% of the incident light.

Nonequilibrium carriers were excited by a cw Ar^+ laser with a power up to 3 W. The intensity of the laser light incident on the crystal was held constant during an experiment; the variations in this intensity were less than 5%. The luminescence was measured by a lock-in detection method. The 5145-Å laser light was modulated at a frequency on the order of 30 Hz. The spectrum was analyzed with the help of a diffraction monochromator with a 600-lines/mm grating and a linear dispersion of 26 Å/mm in the working region. The luminescence signal was detected by a cooled Ge(Au) detector with a sensitivity $\sim 10^{-15}$ W/Hz$^{1/2}$ in the region of interest. The magnitude of the strain was determined from the shift of an exciton line. From changes in the shape and half-width of this line we drew conclusions about the extent of nonuniformity of the strain; it did not exceed 5% at a pressure of 2.5 kbar. The magnetic field was parallel to the compression axis.

Figure 1 shows luminescence spectra of a strained Ge $\langle\sim 100\rangle$ sample during con-

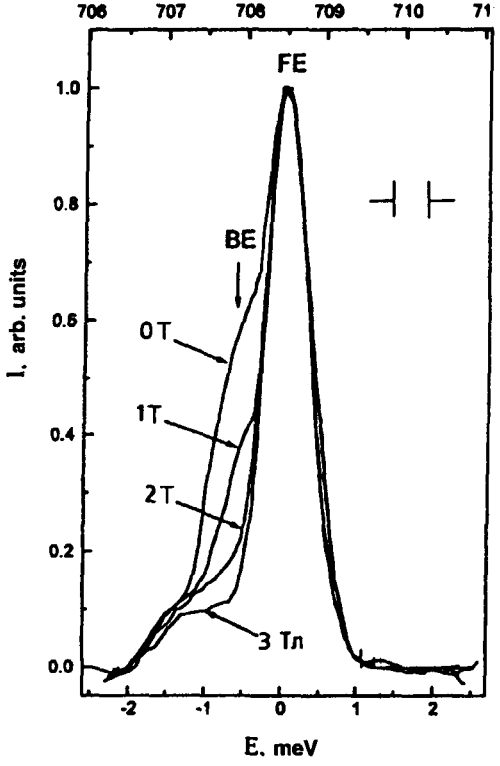

FIG. 1. Luminescence spectra of Ge ⟨~100⟩ with the emission of an *LA* phonon at $P=2.5$ kbar in various magnetic fields. The pump intensity is $I=150$ W/cm^2. The bath temperature is 1.8 T. Knee *BE* corresponds to a biexciton. The values of the energy E on the lower scale are shown in the scale $\hbar\omega-E_0$, where E_0 is the position of the peak of the exciton *LA* line. The upper scale shows absolute values of the energy for the luminescence spectrum in the absence of a magnetic field. The intensities I of the spectra have been normalized at the peak of the exciton line.

tinuous pumping in a field up to 3 T. The knee on the low-energy side of the exciton line grows faster than linearly with increasing pump level; it is suppressed by a weak magnetic field. This result is unambiguous evidence that the knee belongs to a biexciton. A biexciton is destroyed by a field because there is no correction to the biexciton energy which is linear in the field, and a pair of excitons is more favorable than a biexciton from the energy standpoint, because of the orientation of the magnetic moments of the electron and the hole in a field. A secondary cause of destruction of biexcitons is the positive diamagnetic correction to the energy of a molecule in a magnetic field with respect to a pair of unbound excitons. Working from the values of the diamagnetic susceptibility in a weak field,[5] $\chi_{ex}0.036$ meV/T^2 and $\chi_{be}=0.15$ meV/T^2, we find a value on the order of 1.5 T for the field capable of destroying a biexciton. This result agrees with our measurements. From the biexciton knee we can determine the density of excitons, using the condition for thermodynamic equilibrium of excitons and biexcitons ($\mu_{be}=2\mu_{ex}$):

$$n_{be}=n_{ex}^2\left(\frac{4\pi\hbar}{M_{ex}k_BT}\right)^{3/2}\left(\frac{\nu_{be}}{\nu_{ex}^2}\right)\exp\left(\frac{\Delta}{k_BT_{ex}}\right).$$

Here n_{ex} and n_{be} are the densities of excitons and biexcitons, respectively, and M_{ex} is the translational mass of an exciton, which is $\cong 0.35m_0$, where m_0 is the mass of a free electron. We take the biexciton binding energy $\Delta = 0.27$ meV from Ref. 5. The degrees of degeneracy of the excitons and biexcitons are $\nu_{ex} = 4$ and $\nu_{be} = 1$, respectively; T_{ex} is the temperature of the exciton gas. The integral luminescence intensities and the densities of excitons and biexcitons are related by[6] $I_{be}/I_{ex} = 2.3n_{be}/n_{ex}$. The maximum exciton density which we achieved was 0.4×10^{15} cm^{-3} ($r_s = 5.2$). The exciton temperature is estimated to be 3 K on the basis of an approximation of the shape of the exciton luminescence line by a Boltzmann distribution. A further increase in the pump density leads to a heating of the exciton system and to a thermal dissociation of molecules.

In a magnetic field of 5 T, the spin degeneracy of the excitons is completely lifted, and the splitting of the nearest spin levels exceeds the temperature by a factor of 2. Consequently, in highly strained Ge $\langle \sim 100 \rangle$ in a strong magnetic field we are dealing with a gas of excitons in a single nondegenerate state.

Figure 2a shows the position of the exciton LA line versus the square of the magnetic field for two slightly different pressures. The shift of the line in a field is described well by a quadratic law. The diamagnetic susceptibilities turn out to be 0.034 and 0.04 meV/T^2 for pressures of 2 and 2.5 kbar, respectively. These figures are an order of magnitude larger than the susceptibility in a weak field as calculated in Ref. 7 and confirmed experimentally in Ref. 5. One might expect changes in the behavior of the exciton energy as a function of the magnetic field in fields above 1 T, since even at a field of 6 T the diamagnetic correction to the exciton energy is greater than Ry. However, the observed behavior of the exciton shift turns out to be quite unexpected: A pronounced change in χ_{ex} occurs in an interval of fields on the order of 1 T; beyond this interval, there is essentially no change in the shift. The increase in χ_{ex} with the pressure is attributed to a decrease in the mass of the holes, which depends on the magnitude of the level splitting in the valence band. The inset in Fig. 2a illustrates the increase in the exciton binding energy in a magnetic field. The straight line corresponds to the lower spin-split Landau level. The cyclotron mass of the holes is taken in the infinite-pressure approximation. We see that the exciton binding energy increases by a factor of 5 in a field of 14 T.

In addition to the increase in the binding energy, there are changes in the distribution of the electron–hole density in an exciton and the volume of the exciton in a strong field. The compression of the exciton wave function results in an increased probability for radiative recombination, which is inversely proportional to the effective volume of an exciton. In the limit of ultrastrong fields, the probability for radiative recombination of an exciton has a field dependence $1/a_B l_H^2$ [i.e., it is proportional to $H/H_c \ln(H/H_c)$], while the dependence in a weak field is $\sim H_2$ (Ref. 8). This effect should lead to precisely the same increase in the integral intensity of the exciton line under the assumption that nonradiative recombination, which is the fastest mechanism for exciton annihilation, is independent of the field. Under these conditions we would expect a quadratic field dependence of the exciton line intensity in weak fields and an approximately linear dependence in strong fields. Figure 2b shows the intensity of the exciton line during continuous pumping as a function of the square of the magnetic field. In weak fields the increment in the intensity is described satisfactorily by a quadratic law. In stronger fields, we see a "saturation," which we attribute to a transition into the strong-field region.

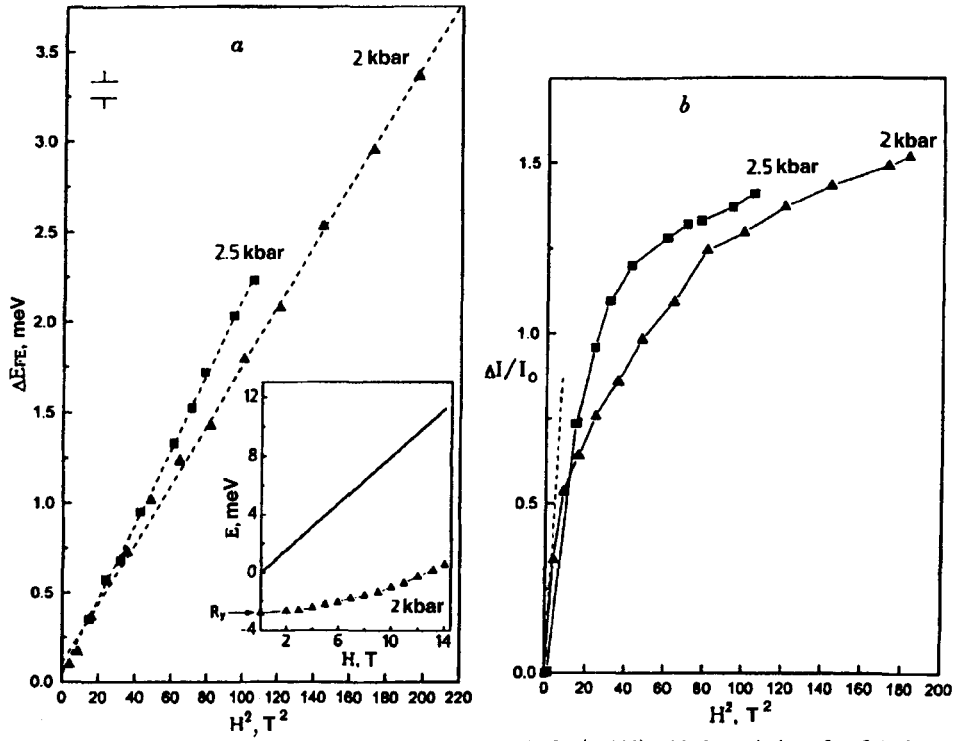

FIG. 2. a: The shift (ΔE_{FE}) of the exciton luminescence line in Ge $\langle \sim 100 \rangle$ with the emission of an LA phonon versus H^2 for two pressures. The bath temperature is $T = 1.8$ K. The inset shows the change in the exciton binding energy in a magnetic field at $P = 2$ kbar. The straight line here is a plot of $\hbar\omega_c/2 - \mu_B H/2(|g_e| + |g_h|)$, where $g_e = 1.57$, $g_h = -4.5$ (Ref. 5), and ω_c is calculated for the masses $m_c^e = 0.135 m_0$ and $m_c^h = 0.109 m_0$. b: Increase in the integral intensity I of the LA exciton luminescence line in a magnetic field. The dashed line is a straight line corresponding to the intensity increase in the case of a weak field:[8] $\Delta I/I_0 = (11/3)(\hbar\omega_c/4Ry)^2 = 0.11$, where I_0 is the integral intensity of the line in the absence of a field.

In a field above 4 T, with strong pumping, a new line, X, appears on the red side of the exciton line. The inset in Fig. 3 gives an idea of the evolution of this line in a magnetic field. As the field is strengthened, the peak of line X shifts away from the exciton line in the red direction, and the width of this line increases. In a field of 10 T, the integral intensity of the line amounts to 15% of the exciton intensity. The formation of a line on the low-energy side of the exciton line implies the formation of states with a binding energy per e–h pair which is larger than that of an exciton. There are two ways to explain the formation of this line. The first is based on the increase in the stability of an electron–hole liquid in a strong magnetic field,[9] although in our case, in contrast with the model proposed in that previous study, the electrons and the holes should be not exclusively in the lower Landau level. In a field of 4 T, the width of line X is on the order of 3 meV. On this basis we estimate the density of the electron–hole liquid to $\sim 10^{16}$ cm^{-3}, and we find the parameter r_s to be $r_s = (4\pi a_{ex}^3 n/3)^{-1/3} > 1$, where a_{ex} is the first Bohr radius of an exciton in the absence of a field. For an electron–hole liquid in unstrained Ge we would have $r_s \cong 0.5$. When the compression of the exciton wave func-

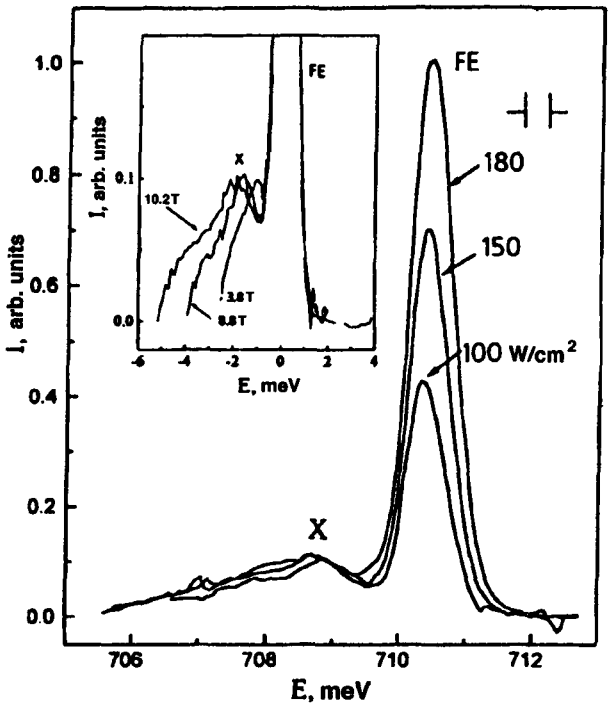

FIG. 3. Behavior of the X line at various pump intensities in a magnetic field of 10.2 T. The bath temperature is $T=1.8$ K. The inset shows line X at a constant pump intensity $I=100$ W/cm^2 in various magnetic fields. The energies are expressed in the scale $\hbar\omega-E_0$, where E_0 is the position of the peak of the exciton LA line. $T=1.8$ K, $P=2.5$ kbar. The spectra have been normalized at the peak intensity of the exciton line.

tion by the magnetic field is taken into account, the parameter r_s increases by a factor of at least 2; i.e., we are dealing with a formation which is rather "porous" in density. It is not clear whether an electron–hole liquid is stable at such values of r_s.

An alternative explanation of line X is based on an increase in the quadrupole interaction between excitons in a magnetic field. The strengthening of this interaction could in principle stabilize a triplet biexciton[1,2] or more-complex molecular formations. A stabler triplet exciton might arise as the result of a quadrupole–quadrupole interaction under the strong inequality $(l_H/a_{ex})(\mu/M)\ll 1$. Under our experimental conditions, the expression on the left side of this inequality is 1/40.

A comprehensive explanation of line X will require experiments at higher pressures (5–7 kbar), since an electron–hole liquid is extremely sensitive to a change in the mass of the holes, while the stability of molecular formations should depend far more weakly on this parameter.

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