Bose gas of spin-oriented excitons in uniaxially deformed germanium

I. V. Kukushkin, V. D. Kulakovskiĭ, and V. B. Timofeev
Institute of Solid State Physics, USSR Academy of Sciences

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A narrowing of the spontaneous, indirect exciton-annihilation line with increasing density of optical excitation has been detected in uniaxially compressed Ge crystals in a magnetic field that orients the spin of the electrons and holes in the excitons. The observed changes in the exciton-phonon radiation spectrum of the high-density exciton gas are explained in terms of the nonideal Bose gas.

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Since excitons have an integer spin, they must obey Bose-Einstein statistics. It has been predicted that the quantum-statistical properties of excitons should be evident in the high-density quasiequilibrium gas at sufficiently low temperatures.[1-3] Certain difficulties arise when it is attempted to realize these conditions experimentally. In the indirect Ge and Si semiconductors the primary difficulty is attributed to the fact that at low temperatures the chemical potential μ_{FE} and the density n_{FE} of exciton gas are limited by condensation into an electron-hole liquid (EHL).[4,5] In unstrained Ge[6] and Si[7] the exciton gas remains classical ($|\mu_{FE}|/kT \gg 1$) along the phase-equilibrium boundary with the EHL to the region in which their ionization breakup occurs. The stability of the EHL can be reduced and therefore the phase-equilibrium boundary can be shifted toward higher exciton-gas densities at low temperatures if the orbital degeneracy in the electron and hole bands is eliminated by means of elastic uniaxial strain.[8] The most suitable conditions are realized in the case of Ge for a deformation along a nonsymmetrical direction close to ⟨100⟩ (Ge ⟨~100⟩) when the EHL binding energy is reduced to ~5 K. However, the exciton molecules can also be formed in this case because of the fairly strong attraction between the excitons. The situation changes radically if Ge⟨~100⟩ is placed in a magnetic field that orients the spins of the electrons and holes in the excitons. The interaction between such "spin-oriented" excitons at close separations has a repulsive character, which results in the breakup of exciton molecules.[10] Because of the weak attraction between excitons at large distances, the stability of the EHL is preserved, although its binding energy is reduced markedly in magnetic fields $H \sim 4$ T. This gives rise to the possibility of studying the statistical properties of exciton gas at $T \gtrsim 1.5$ K and at densities up to $n_{FE} \sim 3$–5×10^{15} cm^{-3} ($|\mu_{FE}|/kT \sim 1$).

We have investigated Ge crystals with a shallow impurity concentration $\lesssim 5 \times 10^{11}$ cm^{-3}. The uniaxial compression method for the 3 × 3 × 10-mm crystals was described previously.[9] Optical excitation was provided by an LT-2 laser ($\lambda = 1.06$ μm) with a power of 4 W. The direction of the magnetic field coincided with the compression axis of the crystal when the observations were made in the perpendicular direction (Voight geometry). We used a double monochromator with a 600-line/mm grating and an 8-Å/mm dispersion. The radiation was detected by a cooled Ge(Cu) photoresistor that operated in the synchronous-detection mode. The spectra were recorded with a resolution of 70 μeV.

The inset in Fig. 1 shows the LA component of the emission spectrum of Ge compressed along an axis that deviates by 3° from the ⟨100⟩ direction at $T = 1.75$ K in a magnetic field $H = 4$ T. At such fields the magnetic length in Ge is comparable to the Bohr radius of the exciton. The magnitude of the deformation 250 MPa ensured complete elimination of orbital degeneracy in the bands. The emission lines of free excitons from the ground spin state (FE) and from the EHL (L) are observed in π polarization. We note that under the conditions of our experiment the spin-excited state of the excitons, which is observed in the σ component of the spectrum, is less populated than the ground state by almost an order of magnitude.

Figure 1 shows the exciton-emission spectra measured in π polarization at bath temperatures $T_b = 2.15$ and 1.75 K and different densities of the exciting light that differ by about an order of magnitude. The magnitude of the smallest paramagnetic splitting in the exciton ($g_e \mu H$) in a field $H = 4T$ is also shown here. To facilitate comparison, the spectra have been normalized to the intensity at the maximum. It can

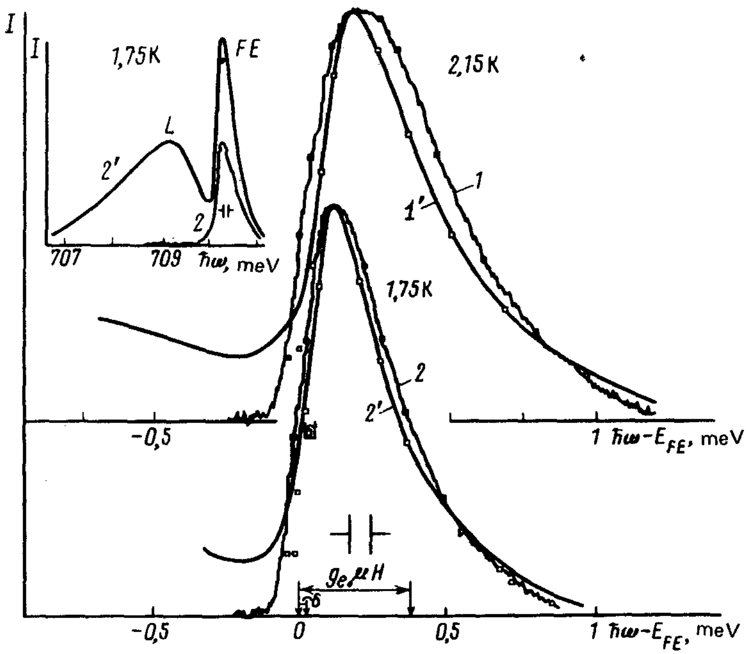

FIG. 1. Exciton luminescence spectra of Ge ⟨~100⟩ with emission of an LA phonon at $H = 4$ T, $T_b = 2.15$ K and 1.75 K, and different excitation densities: 1–5 W/cm², 1'–60 W/cm², 2–4 W/cm², 2'–40 W/cm². Approximation of the spectrum shape within the framework of the Boltzmann (■) and Bose (□) distributions. The complete emission spectra at $T_b = 1.75$ K are shown in the inset.

be seen that the *FE* emission line narrows markedly with increasing density of the gas phase as the excitation power is increased.

Figure 2 illustrates the variation of the half-width of the *FE* line as a function of its intensity and, consequently, of the exciton concentration. The value of γ was determined at the half-height of the contour. The emission line of the excitons from the spin-excited state (σ component) is broadened as the pumping is increased; this indicates that the exciton temperature T_{FE} increases. Nevertheless, despite the increase of T_{FE}, the π component of the *FE* line narrows with increasing n_{FE}, and it begins to broaden only at the highest pumping levels. In our view, the narrowing of the *FE* line with an increase in pumping is direct evidence of the quantum statistics of excitons and is attributable to their integer spin.

We shall analyze the shape of the exciton-phonon emission line. At the minimum excitation densities (~4 W/cm²) the line shape is described well if we assume that the excitons have a Boltzmann distribution in the band. Figure 1 shows the approximation of the line contour by the expression

$$I(E) \sim \sqrt{E}\,\exp(-E/kT) \quad (1)$$

with allowance for the finite width of the spectrometer slits. We emphasize that the distortion of the line by the spectral slit, which did not exceed $(1/5)\gamma$ under the conditions of the experiment, was small. The only adjustment parameter was the temperature of the exciton system. T_{FE} was not drastically different from the bath temperature T_b, and the overheating increased as T_b approached the λ point ($T_{FE} = 2.1$, 2.5, and 3.1 K at $T_b = 1.75$, 1.9, and 2.15 K, respectively). The narrowing of the *FE*

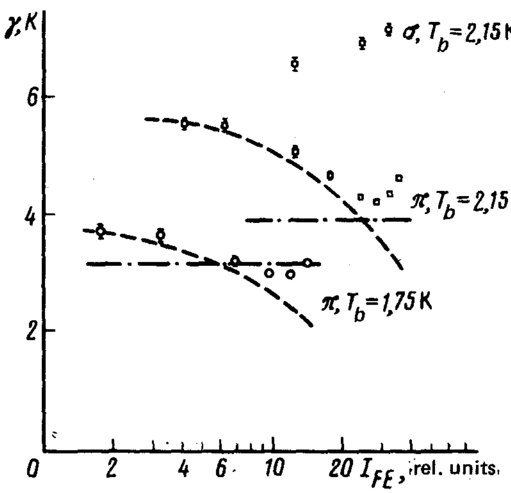

FIG. 2. Dependence of the width of the exciton-phonon luminescence spectrum, measured in π and σ polarizations, on the intensity of the exciton emission line (for pumping interval from 4 to 100 W/cm²). The dashed curve shows the behavior of the spectrum half-width with an increase in exciton concentration within the framework of the ideal Bose gas. The dot-dashed curve shows the predicted spectrum width for a Boltzmann distribution for the condition $T_{FE} = T_b$.

line as n_{FE} increased amounted to 20%. We emphasize that at $T_b = 1.75$ K and a high excitation density the FE line was narrower than that predicted by Eq. (1) even for $T_{FE} = T_b$ (Fig. 2). At such excitation densities the shape of the FE line is described well by the expression (Fig. 1)

$$I(E) \sim \sqrt{E}\left(\exp \frac{E - \mu_{FE}}{kT} - 1\right)^{-1}, \tag{2}$$

which takes account of the Bose distribution of the excitons in the band. By assuming that the Bose gas of excitons is ideal and in quasiequilibrium, we can easily determine μ_{FE} and then the density n_{FE} from the shape of the exciton-phonon line. This method gives a low estimate for the density, since the warming of the excitons with an increase in the pumping, which has been ignored by us, contributes to a boradening of the line. From the spectra shown in Fig. 1 we determined that $\mu_{FE} \approx -0.19$ meV for both temperatures, whereas $n_{FE} = 2.8 \times 10^{15}$ cm^{-3} at $T_{FE} = 3.1$ K and $n_{FE} = 1.2 \times 10^{15}$ cm^{-3} at $T_{FE} = 2.1$ K. The limit for μ_{FE} is evidently determined by condensation into a liquid. By using larger strains (up to 500 MPa) we achieved a further decrease in the liquid binding energy. However, the smallest half-width of the FE line at $T = 1.75$ K remained equal to ~3 K.

It is natural to assume that the exciton gas is nonideal at such high densities, since the dimensionless parameter in this case is $na_0^3 \sim 0.1$, even if the scattering length is assumed to be $a_0 \sim 2a_B$, where a_B is the Bohr radius of the exciton. We recall that $na_0^3 \sim 0.23$ for the highly nonideal He^4 system. It can be assumed that the nonideal nature of the Bose gas of excitons is manifested spectroscopically as a broadening of the FE emission line. The problem of a nonideal Bose gas can be solved theoretically by assuming that a large number of particles is present in the condensate.[11] There is no Bose condensation in the case under consideration. Nevertheless, if we assume that the minimum line width γ in our experiments is approximately equal to the parameter $\Gamma = 4\pi h^2 na_0/M$, which characterizes the width of the energy distribution of a nonideal gas, then we can estimate the scattering length a_0. It turned out that with this assumption a_0 is approximately equal to the diameter of the exciton spheres, $a_0 \sim 2a_B$.

Thus, the gas of spin-oriented excitons in Ge ⟨~100⟩, which at $T \sim 2$ K and $n_{FE} \gtrsim 2 \times 10^{15}$ cm^{-3} is a nonideal Bose gas, is a new, nontrivial quantum object.

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