Condensation of bulk excitons on a magnetized two-dimensional electron gas in modulation-doped heterojunctions

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The evolution of photoluminescence (PL) spectra with increasing magnetic field ($B \leqslant 7$ T) was studied in high-mobility, wide GaAs/$Al_{0.3}Ga_{0.7}As$ heterojunctions (HJ's) at lattice temperatures $T_L = 1.9$–25 K. The two-dimensional electron-gas (2DEG) density in the studied samples is $n_{2D}^0 = (0.9$–$3) \times 10^{11}$ cm^{-2}, and it was varied with He-Ne laser illumination by optical depletion. For $B=0$ the PL is completely dominated by exciton recombination in the undoped GaAs layer. As B increases this band shows a typical exciton diamagnetic shift. For a filling factor $\nu \leqslant 2$ a strong PL transformation is observed: the exciton PL intensity decreases and a new, low-energy PL line abruptly appears and gains intensity at the expense of the exciton PL. We attribute this line to the 2DEG-free hole recombination, and propose that its appearance in the HJ's results from an increased free-exciton dissociation near the 2DEG at $\nu \leqslant 2$. Thus, the evolution of the bulk free exciton to 2DEG-free hole PL with increasing B is due to "condensation" of the bulk excitons on the magnetized 2DEG layer at $\nu \leqslant 2$.

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The unique properties of a two-dimensional electron gas (2DEG) formed in a semiconductor heterostructure are widely studied by photoluminescence (PL) spectroscopy.[1–3] Intrinsic, low-temperature PL resulting from the radiative recombination of the 2D electrons with photoexcited free holes, proved to be an effective optical probe of the many-body interactions and their modification under an applied magnetic field B.[1–8] The PL spectrum, intensity, and decay time have been shown to strongly depend on the 2DEG-hole (e-h) separation,[4–7] and various PL spectral anomalies near a filling factor $\nu \approx 1$ were recently observed in modulation-doped quantum wells (QW) where the e-h separation is determined by the QW width or by the applied bias voltage.[4,6] Many optical studies were done on single heterojunctions (SHJ's) where the GaAs layer was intentionally p-type δ-doped in the vicinity of the 2DEG, and the 2D electron recombines with the hole bound to an acceptor.[1] The resulting inhomogeneous broadening of this *extrinsic* PL line can smear out the effects due to electron-electron or electron-hole interactions in such SHJ's.

In high-quality GaAs/AlGaAs SHJ's without p doping (such as shown in the inset of Fig. 1), the photoexcited electron-hole pairs are rapidly separated by the built-in SHJ electric field spreading over the entire undoped GaAs layer (width >100 nm).[12] The 2DEG-free hole separation becomes very large, and the *intrinsic* 2DEG-hole PL cannot be observed. Indeed, at $B=0$ and $T_L > 2$ K the PL spectrum of such SHJ's is dominated by a narrow PL band at $E \simeq 1.515$ eV originating from free bulk exciton recombination in the GaAs layer.[9–11] It should be noted that some authors attributed this band to the recombination of a hole and the 2D electron from the second subband.[5,14–16] However, under high B applied along the sample growth axis, the magnetophotoluminescence shows a behavior specific of a recombination between a hole and 2D electrons from the lowest subband.[5,13,14,16] These contradictions lead to speculations on the nature of the SHJ PL within the entire range of B. Recently, new features of the magneto-PL spectra have been reported and interpreted as due to an increased overlap of the 2D electron and photoexcited hole wave functions with increasing magnetic field.[15,16] Thus, an understanding of the PL nature and transition from the bulk exciton to the 2DEG-hole PL with increasing B as well as a study of many-body effects on the intrinsic PL in the heterojunctions is required.

We have performed a detailed magneto-PL study for a series of high-quality, single-side modulation-doped

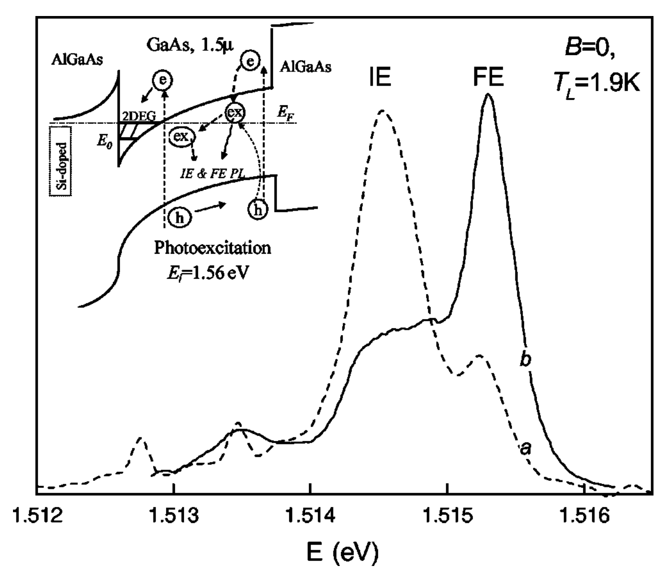

FIG. 1. PL spectra of a wide GaAs/AlGaAs heterojunction observed at $I_L=0$ (a) and 0.4 mW (b). The bands and the processes in the photoexcited HJ are schematically shown in inset.

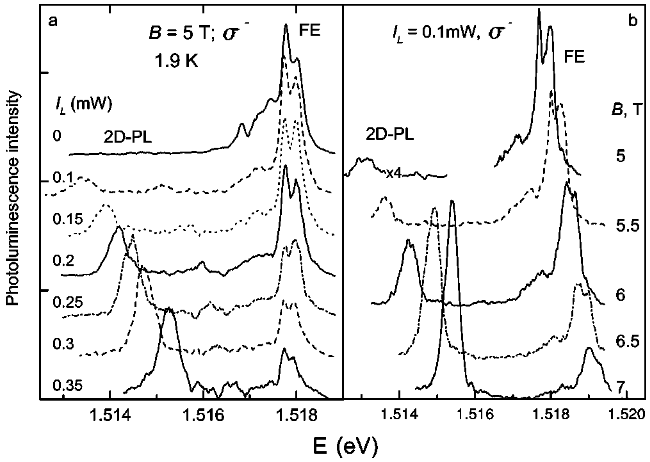

FIG. 2. (a). PL spectra at $B=5$ T for increasing I_L values. (b). PL spectra at $I_L=0.1$ mW and for several B values. The spectra are vertically shifted for clarity.

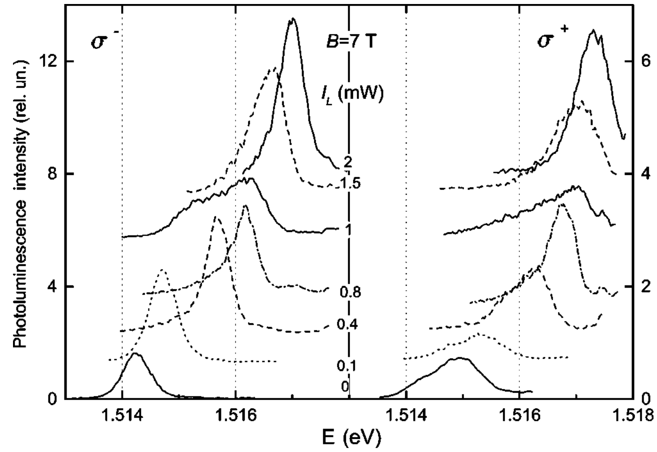

FIG. 3. The PL-spectra recorded at 7 T in two light polarizations with different optical bias ($T=1.9$ K). The vertical scales are different for σ^- and σ^+ polarizations.

GaAs/$Al_{0.3}Ga_{0.7}As$ heterojunction (HJ) structures with a variable 2DEG density (by cw optical depletion[12]). The undoped GaAs layer (having widths of 1–1.5 μ) was separated from a Si-doped $Al_{0.3}Ga_{0.7}As$ layer by a $Al_{0.3}Ga_{0.7}As$ As spacer of (30–70) nm width. The 2DEG density and the dc mobility, measured at $T_L=4$ K for the various samples, are $n_{2D}=(0.9–3)\times 10^{11}$ cm^{-2} and $\mu_{dc}=(1-4)\times 10^6$ $\mathrm{cm^2/V\,sec}$, respectively. An external magnetic field B (up to 7 T) was applied perpendicularly to the HJ plane.

Two light beams from a chopped Ti-sapphire at $E_i=1.56$ eV and a cw He-Ne lasers illuminate the sample. The light intensity of the former was $I_i=(1–5)$ $\mathrm{mW/cm^{-2}}$. The intensity of the defocused cw He-Ne beam (optical bias) was varied in the range $I_L=(0.05–10)$ $\mathrm{mW/cm^2}$. The n_{2D} values under illumination were estimated from dimensional magnetoplasma resonance (DMPR) measurements performed on the mesa samples:[11,17] n_{2D} decreases by a factor of 2–3 within the used range of I_L. The modulated PL spectrum was measured by a standard spectroscopic technique. The PL evolution with increasing B was studied in the same sample for various n_{2D} as controlled by I_L variation. All the studied samples show similar PL spectra at $B=0$ and a similar evolution with increasing B. We discuss the experimental results for the HJ having $n_{2D}=2.6\times 10^{11}$ cm^{-2} under low-intensity Ti-sapphire laser illumination. The sample temperature was varied in the range of $T_L=1.9–25$ K.

At $B=0$, the PL spectra observed for $I_L=0$ and 0.4 mW are presented in Fig. 1 (curves *a* and *b*, respectively). The PL spectrum at $I_L=0$ consists of a bulk free-exciton (FE) band and a low-energy band due to interface-exciton (IE) recombination.[9–11] Their relative intensities vary from sample to sample, with the FE intensity increasing under He-Ne laser illumination (curve *b*) or at higher T_L. It was shown[11] that the PL redistribution as well as a superlinear FE PL intensity increase with I_L, are due to a variation of the built-in electric field when n_{2D} decreases by optical depletion.

The evolution of the σ^- polarized PL spectra with increasing I_L for $B=5$ T is shown in Fig. 2(a). The $I_L=0$ spectrum consists of bands due to FE and localized excitons. The PL intensity of the latter depends on the sample quality. As I_L increases a new low-energy PL band (2D-PL) arises. Increasing I_L causes a blue shift and an intensity enhancement of the 2D-PL band while the FE-PL intensity decreases. The PL evolution with increasing B at $I_L=0.1$ mW is shown in Fig. 2(b). One can see that the 2D-PL band shifts to higher energies faster than the FE-PL band does, and the 2D-PL intensity increases while that of the FE PL decreases. The behavior of both circular polarized 2D-PL bands (σ^- and σ^+) with increasing He-Ne laser intensity at $B=7$ T is shown in Fig. 3. At 7 T and $I_L=0$ the 2D-PL band is already observed; increasing I_L results in the 2D-PL blue shift. At $I_L\approx 1$ mW, both σ^- and σ^+ 2D-PL specta exhibit a large spectral broadening. Below, we will see that $\nu\simeq 1$ at $I_L\approx 1$ mW.

The peak energy of the FE and 2D-PL bands as a function of B is plotted in Fig. 4(a) for three n_{2D} values. The FE-PL energy shows a diamagnetic shift that does not depend on n_{2D}. The FE-PL bands in both σ^- and σ^+ polarizations are split into two lines (see Fig. 2) as is observed for the bulk FE exciton.[9] The 2D-PL energies shift linearly with B, however, these shifts occur faster at low B till the 2D-PL intensity grows. Figure 4(b) displays the evolution of the integrated intensities J of both FE and 2D-PL bands with B. At a certain B value, the 2D-band PL abruptly appears and its intensity sharply grows while the FE-PL intensity decreases. The threshold of the 2D-PL appearance shifts to lower B as n_{2D} decreases by optical depletion.

The temperature dependence of the PL spectra for a given I_L is demonstrated at $B=5$ T in Fig. 5(a); the dependence $J(T_L)$ is plotted in Fig. 5(b). As T_L increases the 2D-PL integrated intensity decreases while the FE PL increases, and the 2D-PL band disappears at 20 K. Thus, as B or T_L varies, a PL intensity redistribution between the FE-PL and 2D-PL channels is observed.

The characteristic feature of the photoexcited SHJ having an undoped GaAs layer, is the coexistence of free excitons in the GaAs layer and a 2DEG near the HJ interface. Free electron-hole pairs are photogenerated practically homoge-

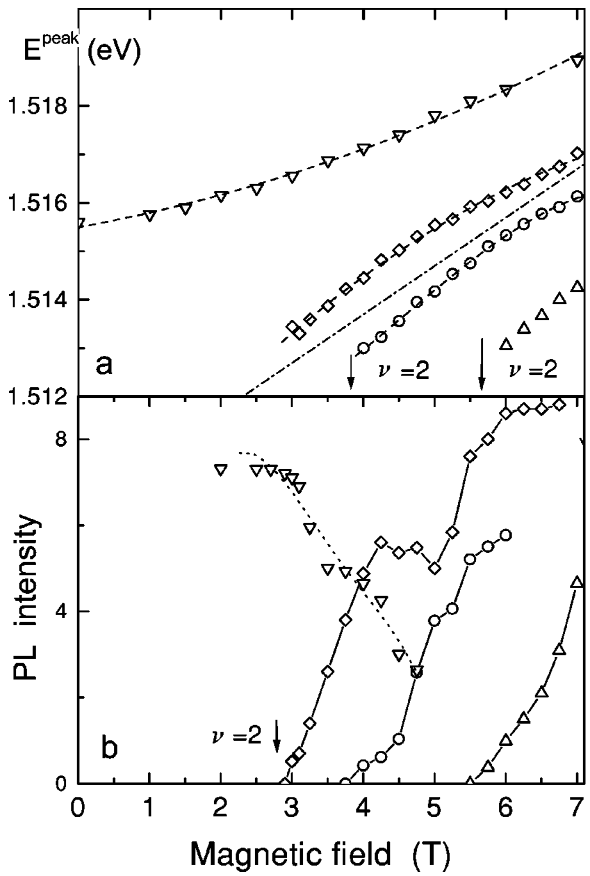

FIG. 4. The peak energy (a) and integrated intensity (b) dependencies on B for the exciton (∇) and for the 2D e-h PL. n_{2D}: $\triangle$ -2.6; $\bigcirc-1.8$; $\diamond-1.3\times10^{11}$ cm^{-2}. The dot-dash line in (a) shows a linear dependence $E(B)$: $E=1.509+0.001B$.

neously throughout the structure since the light penetration length is about 1 μ. Then, the electrons and holes are rapidly separated by the built-in electric field: the holes drift away from the HJ and the electrons drift towards the 2DEG. In the flat band region of the GaAs layer, electrons and holes bind into excitons that diffuse and drift to the higher-field region (see inset of Fig. 1).

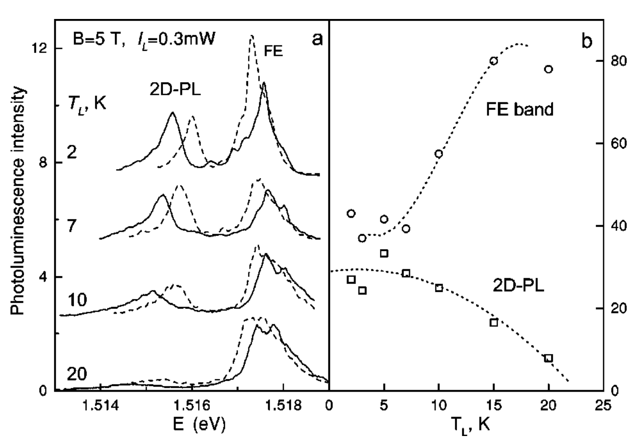

FIG. 5. (a) PL spectra for $B=5$ T at various temperatures. (b) The integrated intensity of both PL bands vs T_L ($I_L=0.1$ mW).

In thick GaAs layers having a high-electron mobility at low temperature,[18] the exciton diffusion length $L=(D\tau_{ex})^{1/2}\simeq5\times10^{-3}$ cm exceeds the undoped GaAs layer width, and the FE density is controlled by the boundary conditions, namely, by the rate of exciton recombination or dissociation at the AlGaAs-GaAs and GaAs-AlGaAs interfaces. Here, we use an exciton diffusion coefficient $D\simeq100$ cm^2/sec and an exciton lifetime $\tau_{ex}\simeq3\times10^{-9}$ sec, respectively.[19]

Under low B, two PL channels originating from the free excitons, should be considered: (a) GaAs free excitons and (b) Interface excitons. The latter are excitons driven by the electric-field gradient to the 2DEG and situated at the distance of 3–5 Bohr radii from the 2DEG. There, a shallow potential minimum for the translational exciton motion is formed due to the joint effect of the built-in electric field and repulsive short-range forces at the interface.[10]

The low-energy PL band emerging at higher B, is unambiguously attributed to a free hole-2D-electron recombination from the lowest 2DEG subband. Indeed, this band shows a typical linear $E(B)$ dependence for high B with a slope of 1 meV/T [shown by dashed line in Fig. 4(a)] that corresponds to the standard electron effective mass $m_e=0.068$ m_0 (at $B>3$T) and the free hole mass of 0.045 m_0. The PL band is blue shifted with decreasing n_{2D} due to the decreased band-gap renormalization resulting from the electron-electron interaction. We have estimated the n_{2D} values from the microwave DMPR on the mesa sample. Thus, we obtain the same filling factor $\nu\simeq2$ for the 2D-PL thresholds occurring at different n_{2D} values (varied by I_L). Taking $\nu\simeq2$ for the 2D-PL threshold we find that the strong spectral modifications of the σ^- and σ^+ 2D-PL bands (Fig. 3) occur at the filling factor $\nu\simeq1$. Spectral peculiarities near $\nu\simeq1$ were previously reported for wide QW's, and these were attributed to a skirmion-hole radiative recombination when the lower- and higher-energy spin state is completely full and empty, respectively.[4,6,7] Thus, the effect of changing of the Landau sublevel occupancy on the polarized PL spectra at $\nu\simeq1$ is also clearly observed in the high-quality HJ's, however, in contrast to the QW's case, a large PL line broadening appears in both σ^- and σ^+ polarizations (Fig. 3).

The remarkable threshold appearance of the 2D PL and quenching of the FE PL as well as the reduction of the 2D-PL intensity with temperature (with an activation energy of 1 meV) resemble the phase transition of the excitons into an electron-hole liquid in bulk Ge and Si.[20] We propose that the interaction of the free excitons with the magnetized 2DEG results in an exciton dissociation near the 2DEG. The experimental results lead to the conclusion that at $\nu\leqslant2$, the energy of the excitonic hole-2D-electron system becomes lower than the exciton binding energy, so that the free exciton dissociates into a 2D-electron and free hole. The hole is self-localized in the potential well situated close to the 2DEG, and recombines with the 2DEG. The sharpness of the transformation allows us to suggest that a nonequilibrium phase transition of the excitons takes place near the interface with the magnetized 2DEG at $\nu\leqslant2$. Perhaps, a strong decrease of the 2D-electron Fermi energy at $\nu\leqslant2$ promotes this transition. Recently, a hysteretic behavior of a 2D-PL intensity near $\nu \simeq 2$ has been reported at $T_L < 1$ K,[16] and, it is likely to point out the first order of such a transition. Note, that the transformation of the free excitons into 2D-electron-hole pairs can be phenomenologically considered as a sharp increase of the exciton interface recombination rate as $\nu \leqslant 2$.

At B values close to the threshold, the 2D-PL peak energies increase faster with B than at higher B [Fig. 4(a)]. It is likely to be due to a decrease of the hole-2DEG distance of the formed system as B increases. Since the 2DEG-hole recombination is spatially indirect, this may explain the faster 2D-PL energy shift and the strong PL intensity increase with B.

The PL intensity redistribution at $\nu \leqslant 2$ in HJ's was previously reported.[13,15,16] At low B ($\nu > 2$), the strongest PL band has been attributed to the recombination of the photoexcited hole with the 2D electron from the second confined state at the interface.[5,14–16] This assignment is probably wrong for the following reasons. The photoexcited hole drifts from the interface at the distance of 10^{-5} cm in a very short time of 10^{-10} sec. (Here, we assume a lowest hole mobility—10^3 cm^2/V sec and the lowest HJ electric field—10^2 V/cm). Thus, the 2D-electron-free hole wave function overlap becomes negligible before the free hole-2DEG recombination can occur (the radiative lifetime is larger than 10^{-10} sec). Another reason is the absence of any PL due to recombination of the 2D electron from the lowest subband with a hole, in contrast to its dominance in wide GaAs quantum wells.[4,6] It was speculated that the observed PL-intensity redistribution with increasing magnetic field is caused by an increase of the overlap of the electron-hole wave functions.[13,15,16] However, it is improbable that the holes photogenerated in the wide GaAs layer, far from the interface, will appear near the 2DEG.

The proposed model of ''exciton condensation'' explains the appearance of free holes photogenerated homogeneously in the wide GaAs layer, near the 2DEG. It also explains the transformation of the bulk exciton PL into the low-energy free hole-2D-electron PL with increased magnetic field [Fig. 4(b)] as well as a reverse PL redistribution with increasing ambient temperature [Fig. 5(b)].

In conclusion, a threshold changeover of the free exciton to 2DEG-hole photoluminescence was observed at $\nu \leqslant 2$ in the GaAs/AlGaAs heterojunctions. We propose that the observed phenomenon is due to a ''condensation'' of the bulk free excitons on the magnetized 2DEG layer and an exciton dissociation into 2D electron and hole. A detailed analysis of the exciton-magnetized-2DEG interaction as a function of their separation is needed for the theoretical justification of the observed phenomenon.

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