Transition from two-dimensional electron-hole to geminate-exciton photoluminescence in GaAs/Al$_x$Ga$_{1-x}$As heterostructures under a high in-plane magnetic field

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We studied the evolution of the photoluminescence (PL) spectra in modulation-doped GaAs-based heterostructures (single quantum wells and heterojunctions) at $T = 1.2$ K under a high magnetic field $B$ (up to 33 T), which was applied parallel to the two-dimensional electron gas (2DEG) layer. Under low in-plane fields, $B_{||} < 7$ T, the radiative recombination of the photoexcited hole with the 2DEG gives rise to a broad PL band that shifts quadratically with $B_{||}$. This band transforms into a narrow PL line whose peak energy $E$ shifts linearly with $B_{||}$ in the range of $10^{-33}$ T. The slope of the linear $E(B)$ dependence was measured as $\alpha_E = 0.77 \pm 0.02$ meV/T in all the studied structures. The same linear slope is also measured in the PL spectra of bulk, undoped GaAs under high $B$. We thus attribute the sharp PL line observed in the doped heterostructures to magnetoexcitons that are photogenerated outside the 2DEG layer by a geminate formation process. The slope of the magnetoexciton energy dependence on $B_{||}$ is compared with that measured for unbound-electron-hole Landau level transitions under a perpendicular $B_{\perp}$. The ratio of the measured slopes, $\alpha_E/\alpha_B = 0.8$, is found to be equal to the ratio of the reduced excitonic mass to the reduced cyclotron mass of GaAs.

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I. INTRODUCTION

The radiative recombination of a two-dimensional electron gas (2DEG) with photoexcited free holes in modulation-doped heterostructures [quantum well (MDQW) and single heterojunction (SHJ)] under a perpendicularly applied magnetic field $B_{\perp}$ gives rise to the photoluminescence (PL) (2De-h PL) spectra that contain valuable spectroscopic information on the 2DEG many-body interaction, particularly in the quantum Hall effect regime.1-4 The existing theoretical models for the 2De-h PL consider the recombination of the two-dimensional (2D) electron with a valence hole that is separated by a distance of $d_0$ from the 2DEG layer.5 All the dynamical processes, namely, photoexcitation, space-time evolution of the holes, as well as an exciton formation, are ignored.

The importance of dynamical processes in photoexcited heterostructures containing a 2DEG was manifested in the resonant enhancement of inelastic light scattering by the 2DEG when the photon energy is resonant with quantum-well excitons having energies higher than the Fermi energy.6 Dynamical processes also control the occupation of photoexcited states and give rise to the coexistence of several high-energy PL lines in the low-temperature magneto-PL spectra of MDQWs at electron filling factors $\nu < 1$.7 Another case is a remarkable transformation of the excitonic recombination spectrum in high-quality modulation-doped SHJs to 2De-h PL spectrum that occurs with increasing $B_{||}$ (at $\nu < 2$).8,9 This was explained by the dissociation of the excitons, generated in the thick GaAs layer, at the 2DEG interface with subsequent free hole recombination with the 2D electrons.4 Under low magnetic field applied parallel to the SHJ’s interface ($B_{\perp}$ $\sim$ 1-2 T), 2De-h PL intensity enhancement and spectral modifications were discovered and explained by a free hole spatial redistribution.7

At $B = 0$, an electron and a hole photogenerated at the same point of space (geminate e-h) are instantly separated since they have momenta that are equal in magnitude and opposite in direction. Then, the hole can recombine with a 2D electron or bind an electron by the bimolecular exciton formation mechanism.8 An applied magnetic field strongly affects the motion of the photoexcited charge carriers and their spatial distribution at rather low $B$.3 At high $B$, the exciton binding energy significantly increases.10,11

In the present paper, we report on the effect of a high magnetic field, applied parallel to the 2DEG plane ($B_{||} < 33$ T), on the photoexcited system dynamics which, in turn, affects the PL spectrum of modulation-doped heterostructures. In all the structures studied here, a transformation is observed from a broad 2De-h PL spectrum at low $B_{||} < 7$ T (reported in our previous study7) to a narrow PL line at high $B_{||}$. The peak energy of this line shifts linearly with $B$ above 10 T. The slope of the linear $E(B)$ dependence was measured as $\alpha_E = 0.77 \pm 0.02$ meV/T in all the studied samples. We attribute this PL line to magnetoexcitons that are photogenerated outside the 2DEG layer by a geminate formation process.

The paper is laid out as follows. Section II details the experimental procedure followed (in Sec. III) by the experimental results. In Sec. IV, we discuss the effects of $B_{\perp}$ on the
2De-h PL spectra in MDQWs and SHJs. In Sec. V, geminate-exciton formation under a high magnetic field is considered. The dependence of the magnetoexciton energy on $B_z$ is compared with that of the lowest Landau transitions between conduction and valence subbands under $B_z$ and is analyzed in Sec. VI.

II. EXPERIMENTAL DETAILS

Several MDQW and SHJ GaAs/AlGaAs samples were studied, all grown along the (100) direction by molecular beam epitaxy. The MDQW samples contain a single quantum well of various widths (44, 25, and 20 nm), and the SHJ samples have a thick GaAs buffer layer (width of 1 μm). The $n$-type δ-doped layer is separated from the interface by an undoped AlGaAs layer (widths of 50–100 nm). The 2DEG density range was $n_{2D}=(0.7–2.5) \times 10^{11}$ cm$^{-2}$ and the 2DEG mobility was $\mu=(2–5) \times 10^6$ cm$^2$/V s at 2 K. A Ti:sapphire laser illuminated the sample, and the incident light intensity (photon energy of 1.58 eV) was kept below $10^{-2}$ W/cm$^2$. A single optical fiber was used for photoexcitation and PL collection from the sample. The sample was mounted in a cryostat and was immersed in liquid $^3$He at temperature of $T_L=1.2$ K. The cryostat was placed in an electromagnet producing a magnetic field that reached a maximum value of 33 T. The sample plane (the 2DEG layer) was parallel to $B_z$, while the photoexciting laser beam and the outgoing PL were directed perpendicularly to the sample plane. The PL was measured with a spectrometer equipped with a cooled charge coupled device, and the spectral resolution was $\sim 0.1$ meV.

III. EXPERIMENTAL RESULTS

Figure 1(a) shows a series of PL spectra measured at increasing $B_z$ for the 44-nm-wide MDQW ($n_{2D}=1 \times 10^{11}$ cm$^{-2}$). The panel on the left hand side displays the PL spectra at the low-$B_z$ range. At $B_z=0$, a typical 2De-h PL spectrum of MDQWs is observed. The PL intensity reaches a maximum at the low-energy side ($E_g$), and it vanishes at $E=E_g+E_F$ ($E_F$ is the 2DEG Fermi energy). Thus, the PL bandwidth is of 3.6 meV. As $B_z$ increases to 8 T, the spectrum transforms into an asymmetric PL band with its maximum intensity at the high-energy side showing a blue nonlinear (diamagnetic) shift.

For $B_z>9$ T [right panel of Fig. 1(a)], the PL spectrum is dominated by a narrow line (width $<0.5$ meV) whose peak energy dependence on $B_z$, $E(B_z)$, is plotted in Fig. 1(b) (open circles). It is well approximated by a linear dependence with a slope of 0.79 meV/T (dashed line). The integrated PL intensity ($J$) is also shown in Fig. 1(b) (crosses). It varies little (<25%) in the range of 0–33 T. A similar PL behavior was observed for all the studied MDQWs; however, the $B_z$ values at which the linear $E(B_z)$ dependence starts are higher for the narrower (25 and 20 nm wide) MDQWs.

Figure 2 shows a similar series of the PL spectra obtained with the SHJ sample having $n_{2D}=1.5 \times 10^{11}$ cm$^{-2}$. The main difference between the SHJ and MDQW spectra is clearly seen in the low field range, $B_z<3$ T. At $B_z=0$, the SHJ spectrum consists of two narrow excitonic lines that originate in the 1-μm-thick GaAs buffer layer: the high-energy line is due to free excitons and the low-energy one to localized excitons. The very weak, broad band below the excitons is the PL at $B_z=0$ is very low (in SHJs having $n_{2D}>1 \times 10^{11}$ cm$^{-2}$), because the strong built-in electric field $E_{bj}$ accelerates the holes away from the GaAs/AlGaAs interface where the 2DEG forms.

A remarkable modification of the SHJ PL spectrum upon applying a low in-plane field ($B_z<4$ T) is seen in left panel of Fig. 2. With increasing $B_z$, the 2De-h PL intensity increases while the excitonic PL intensity diminishes. The spectrum transforms into a strongly asymmetric band with its maximum at the high-energy PL side, and the excitonic lines are hardly seen. With further field increase ($B_z>8$ T, right panel of Fig. 2), the PL spectrum narrows and becomes sym-
FIG. 2. PL spectra of a SHJ under in-plane $B$. The low-field spectra (left panel) show a transformation from the excitonic PL to 2De-h PL occurring at $B_l < 3$ T. At higher $B_l$ (right panel), the 2De-h PL band transforms into a geminate-exciton PL line.

Since the SHJs contain a 1-$\mu$m-thick, undoped GaAs buffer layer, we found it beneficial to compare their magneto-PL spectra under $B_l$ with those of a bulk, undoped GaAs layer (10 $\mu$m-thick). Figure 3 (right panel) displays some magneto-PL spectra of this GaAs layer, observed at several $B$ values. At $B=0$, the spectrum consists of free (FE) and bound exciton PL lines, and as $B$ increases, the free exciton line dominates the spectrum. Under high fields ($B > 15$ T), it splits into two lines. The peak energy $E$ dependence on $B$ (of the free exciton line and of the center of gravity for the two split lines) is plotted in Fig. 3 (open circles). This curve is fitted with a quadratic (diamagnetic) exciton shift for $B < 7$ T, and at higher fields, it is well approximated by a linear function with a slope of 0.77 meV/T (solid line).

In Fig. 4(a), we compare representative PL spectra of two SHJs having different $n_{2D}$, measured at $B_l = 6$ and 26 T, with those of the undoped GaAs sample. The SHJ spectra show similar shifts as those of the undoped GaAs sample, but the latter has impurity-bound exciton bands, which are lacking in the former. Also, the PL line of the SHJ with higher $n_{2D}$ is observed at lower energy than that of the lower density SHJ, and both are still lower than the GaAs free magnetoexciton. The $E(B)$ dependence of these three samples is compared in Fig. 4(b). In the low-$B$ range [inset of Fig. 4(b)], the $E(B)$ dependencies for the SHJs follow the diamagnetic shift of free (bound) excitons of bulk GaAs ([at $B < 4$, the FE peak energies coincide for all three samples]. As the excitonic PL lines in the SHJs are transformed into the 2De-h PL band, the PL peak energy in the range of 2-10 T shows a diamagnetic shift, which is weaker than that of the free exciton.

In the range of 10-33 T, the PL line positions of the studied SHJs samples move almost in parallel to that of the bulk FE line. All $E(B)$ dependencies are approximated by straight lines with a slope of 0.77±0.02 meV/T. The energy differences between the $E(B)$ line for the SHJs and the bulk GaAs sample are ~1 meV [for SHJ(a), low $n_{2D}$] and ~2 meV [for SHJ(b), high $n_{2D}$] as seen in Fig. 4(b). It is important to notice that the integrated PL intensity $J$ varies only slightly in the entire range of 0-33 T.

IV. 2De-h SPECTRAL CHANGE UNDER IN-PLANE $B_l < 10$ T

Before discussing the PL spectra that are observed under a high in-plane magnetic field in various modulation-doped heterostructures, we briefly review the physical model of a...
2DEG subjected to a built-in electric field $E_{\text{hj}}^z$ and an in-plane magnetic field $B_{\parallel}$ (see Fig. 5). This crossed field configuration causes an electron drift in the y direction, and it leads to a shift of the electron subband minimum to high wave vectors $k_y B_{\parallel}$ and to an increase in the in-plane electron effective mass (in the y direction).

We reported recently on the effect of an in-plane magnetic field on the 2DE-h PL spectra of heterostructures in the range of $B_{\parallel} < 7$ T. The 2DE-h PL spectral modifications with increasing $B_{\parallel}$ were then explained by invoking direct optical transitions in an indirect band gap structure that results from the shift in $k$ space of the conduction and valence subbands by $k_{\parallel,y}^* = d_0/L_y$ [as shown schematically in Fig. 5(b)]. Here, $d_0$ is the 2DE-h spatial separation and $L_y$ is the magnetic length. In addition, the 2DEG Fermi energy decreases due to the electron effective mass increase. For such an indirect band gap structure, the direct optical transitions giving rise to the same emitted photon energy involve the conduction- (and valence-) band states of various energies (according to the momentum and energy conservation laws). This is in contrast to the optical transitions occurring in the direct band gap structure (e.g., at $B = 0$), and thus, a dramatic modification of the 2DE-h spectrum is caused by applying in-plane $B_{\parallel}$.

In SHJs, the in-plane magnetic field causes spatial redistribution of the photoexcited holes, in addition to the band gap modification discussed above. Electrons and holes (e-h) are photoexcited across the whole 1-$\mu$m-wide undoped buffer GaAs layer. The photoexcited holes drift in the built-in electric field $E_{\text{hj}}$ formed by the positive charged donors in the AlGaAs barrier. Assuming homogeneous e-h photogeneration in the GaAs layer with a rate $G$, a steady-state spatial distribution of free holes along the $z$ axis, $p(z)$, in the presence of the constant $E_{\text{hj}}$, can be obtained from the equation

$$ G - p/\tau + \mu E_{\text{hj}} (dp/dz) = 0, \quad (1) $$

and its solution is as follows:

$$ p(z) = G [\exp(\mu E_{\text{hj}}) - 1]. \quad (2) $$

Here, $\tau$ is the hole lifetime and $L_E = \mu/\tau E_{\text{hj}}$ is the drift length ($\mu$ is the hole mobility). The expected spatial distribution of the itinerant holes is schematically drawn in Fig.
5(a). At large $z$, $p(z)$ decreases since $E_{hj}$ decreases and the binding of e-h pairs into excitons becomes important. It is evident that such a hole distribution results in a low 2De-h PL intensity and an intense GaAs exciton PL as is seen in the PL spectrum shown at $B=0$ in Fig. 2.

As the in-plane field $B_{||}$ increases, the distance between the itinerant photoholes and the 2De decreases. Indeed, the hole exhibits a cycloidal motion along the $y$ axis, and the hole drift in the $z$ direction is suppressed. This results in the hole density redistribution that is schematically shown in Fig. 5(a), and the 2De-h PL intensity increases at the expense of the bulk excitonic PL.

In contrast to the MDQWs, however, the 2De-h PL of SHJ consists of the radiative recombination of the 2De pairs that have various spatial separations $d_{ij}$ and therefore, it is a sum of spectra corresponding to various $k_y^2 = d_{ij}^2/12L_y^2$. Under $B_{||}$, this gives rise to the distinct 2De-h PL spectrum that has a long low-energy tail (for example, as seen in Fig. 2 at 4 T).

At high $B_{||}$ (>8 T), the PL line of both SHJ and MDQW samples significantly narrows [Figs. 1(a), 2, and 4(a)]. The $E$($B$) dependence of this line becomes linear and can be characterized by nearly the same slopes for all the studied samples.

The nature of the narrow PL lines appearing at high $B_{||}$ must be different from the 2De-h recombination. At $B_{||}$ >8 T, the conduction and valence subbands are shifted in $k$ space by a $k_F$ value that exceeds the Fermi wave vector $k_F$, and the highest kinetic energy of the hole participating in the direct optical transition [see Fig. 5(b)], $E_h$ = $\hbar^2(k_x - k_F)^2/2m_h$, increases with $B_{||}$ However, the occupation of these states exponentially decreases if the photoexcited holes have a thermalized Boltzmann distribution near the top of the valence band. Thus, at $T_2 = 1.2$ K, only a few photoexcited, nonthermalized holes can participate in the direct optical transitions with the 2DEG, and the radiative lifetime would greatly increase with $B_{||}$. It would result in a strong PL quenching at high $B_{||}$ in the presence of any nonradiative recombination channels.

A similar PL quenching was observed in the spatially direct exciton PL of double quantum-well structures under an in-plane field. However, no reduction in the integrated PL intensity with $B_{||}$ up to 22 T was recently detected in similar double QW structures. This fact was explained by a better sample quality and introducing a localized exciton recombination process that relaxes the wave vector conservation for the indirect exciton radiative recombination.

In the studied MDQWs and SHJs, the integrated PL intensity does not quench by the high $B_{||}$ [up to 33 T, Fig. 1(b)]. We cannot attribute this to any localization since the radiative recombination of localized holes with the 2DEG would result in a broad 2De-h PL band (as was observed in Ref. 18), in contradiction with the experimental results in Figs. 1 and 2. We explain the observed spectral PL evolution in MDQWs and SHJs under high $B_{||}$ (Figs. 1–4) by a change-over of the 2De-h radiative recombination into the PL of free, geminate-formed excitons.

V. GEMINATE-EXCITON FORMATION UNDER STRONG MAGNETIC FIELD

In the previous discussion of the 2De-h PL modification induced by a magnetic field, it was assumed that an electron and a hole photogenerated at the same point of space (geminate e-h) are instantly separated, and then, the hole can recombine with the 2DEG. The geminate electron and hole moving in opposite directions lose their mutual memory (and thus become unbound) after a time interval of $\theta$. This time can be estimated using the model of electron capture by an attractive center (hole). In this model, the electron is captured if it loses the energy of $kT_2$ being in the sphere of radius $R_T = e/kT_2$ surrounding the hole ($k$ is the dielectric constant).

At $B=0$, the geminate e-h pair will be separated by a distance of $R_T$ for $\theta = R_T/V \sim 10^{-11}$ s (taking a relative velocity of $V \sim 10^7$ cm/s and $T_2 = 2$ K). This time is much shorter than the energy relaxation time due to electron-phonon scattering, $\tau_m = \tau_m(kT_2/4m_t^2) \sim 5 \times 10^{-10}$ s. Here, $s$ is the sound velocity and $\tau_m \sim 4 \times 10^{-11}$ s is the momentum relaxation time for electrons having mobility above $10^6$ cm$^2$/V s. Since $\theta < \tau_m$, the geminate e-h pair does not directly form an exciton and is separated. Thus, the 2De-h recombination takes place before the slow-acting bimolecular exciton formation occurs.

Under a high magnetic field, the geminate-exciton formation process controls the PL of semiconductors. The importance of geminate recombination was considered in several theories, in connection with the observed enhancement of PL intensity and Raman light scattering efficiency under a magnetic field.

We propose that the observed PL spectral evolution under high magnetic fields (Figs. 1–4) is due to a transformation of the 2De-h PL into the PL of geminate-formed magnetoeexcitons.

In the plane perpendicular to the magnetic field, the photogenerated electron and hole move on the same circular orbits and in the opposite directions (or on close cycloids in the case of $E_{hj}^2$ and $B_{||}$). Their separation in this plane occurs by a diffusion process with a coefficient $D = (r_B^2/\tau_m)$, where $r_B$ is the cyclotron radius. The geminate electron-hole pair conserves their mutual memory during a time of $\theta = R_T^2/D = (R_T/r_B)^2\tau_m$. At $B > 5$ T, $\theta$ is of the order of $10^{-11}$–$10^{-10}$ s, and $\theta > \tau_m$. Thus, the geminate electron and hole lose energy and bind into an exciton before they are separated (the geminate pair moving parallel to $B$, in general, will be separated). It is worth noting that the geminate-exciton formation mechanism is most important for polymers and semiconductors having low carrier mobility where geminate pairs are slowly separated even in the absence of $B$.

Thus, we attribute the narrow PL lines emerging in all MDQWs and SHJs under high $B$ to free, geminate-formed magnetoeexcitons. The weak integrated PL intensity dependence on $B$, measured in all the studied samples [Fig. 1(b)] is also explained by the transformation of the indirect in $k$-space 2De-h PL to the geminate-formed free magnetoeexciton PL. This underlines the importance of the dynamical processes in controlling the PL spectrum. Geminate-formed magnetoeexcitons also play an important role in the interpre-


VI. FREE EXCITONS UNDER A HIGH MAGNETIC FIELD

In the range of $B = 10 - 33$ T, all the studied GaAs-based structures exhibit a linear $E(B)$ dependence with the same slope, $\alpha_{ex} = 0.77 \pm 0.02$ meV/T (Figs. 1-4). For the SHJ samples subjected to a perpendicularly applied field $B$, the high-energy PL line (free exciton PL) has an $E(B)$ dependence which is similar to that of $E(B)$, while the low-energy, 2De-h PL line shows a different $E^{2De-h}(B)$ dependence. The latter is determined by the shift of the lowest-energy electron-hole Landau transition $E^{2L}$ with increasing $B$.

In MDQWs subjected to $B$, the main (lowest energy, 2De-h) PL line follows the $E^{2L}(B)$ dependence at low $B$ (at filling factors $\nu > 2$). At higher $B$, (above $\sim 10$ T), exciton-like lines (charged and neutral excitons) dominate the PL spectrum of GaAs-based MDQWs (having $n_{2D} \approx 2 \times 10^{11}$ cm$^{-2}$). Their $E(B)$ can also be approximated by a nearly linear dependence that has the same slope of 0.77 ± 0.02 meV/T.$^{3,26}$

Such a universal linear $E(B)$ dependence of the geminate-formed exciton PL line observed at high $B$ deserves special consideration. Its slope $\alpha_{ex}$ is likely to be an important inherent parameter which is determined by the $B$ dependence of the exciton binding energy $\delta E_x(B)$, since $E(B) = E^{2L}(B) - \delta E_x(B)$.

For an unbound electron-hole pair, the energy of the optical transitions between the lowest Landau levels of the conduction and valence subbands varies as

$$E^{2L}(B) = E_{go} + (1/2) (\hbar e/c \mu_e) B,$$

where $E_{go}$ is the band gap energy at $B = 0$ and $\mu_e = m_e m_h/(m_e + m_h)$ is the reduced electron-hole cyclotron effective mass. In the approximation of simple, parabolic conduction and valence bands, the slope of the linear $E^{2L}(B)$ dependence is $\alpha_{ex} = (1/2) (\hbar e/c \mu_e) = 0.96$ meV/T for electron and hole cyclotron effective masses of bulk GaAs, $m_p = 0.066 m_0$ and $m_s = 0.52 m_0$. For a 20-nm QW, $\alpha_{ex} = 1.02$ meV/T (for $m_s = 0.35 m_0$).

The dotted line in Fig. 1(b) shows the $E(B)$ dependence of the lowest electron-hole Landau level transition for the 44-nm MDQW. From this line, we get $\alpha_{ex} = 0.96$ meV/T, and thus $\mu_e = 0.06 m_0$. This determines the hole cyclotron mass value of 0.02 $m_0$.

The obtained $E(B)$ dependence for the geminate-formed exciton PL suggests that the magnetoexciton binding energy $\delta E_x$ depends almost linearly on $B$ in the range of $10 - 33$ T. In the range of $B$ and $\gamma$ the $E^{2L}(B)$ dependence was analyzed in several theoretical approximations.$^{10}$ For simple band structures, these theories do not predict a linear $E(B)$ dependence.

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magnetoexcitons is characterized by a universal slope of \( \alpha_{ex}/\alpha_0 = 0.77 \pm 0.02 \, \text{meV/T} \). It is different from that of unbound-electron–hole Landau level transitions under a perpendicular \( B \). The ratio of these slopes (\( \alpha_{ex}/\alpha_0 = 0.8 \)) is equal to the ratio of the reduced excitonic mass to the reduced cyclotron mass, \( \mu_e/\mu_c \) of GaAs. We show that two simple, empirical expressions based on three well-known GaAs parameters \( \mu_0 \), \( \mu_e \), and \( \kappa \), allow us to describe the magnetoexciton energy dependence in the entire \( B \) range.

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27. At 30 T, the lowest electron energy (0 Landau level) shifts by ~25 meV, and \( m_e \) increases by <5% due to conduction-band nonparabolicity. Such a nonparabolicity can be taken into account by a slight slope change (by 0.02 meV/T) for the studied \( B \) range.