Magneto-optics of the spatially separated electron and hole layers in GaAs/Al$_x$Ga$_{1-x}$As coupled quantum wells

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We report on the magneto-optical study of the spatially separated electron and hole layers in GaAs/Al$_x$Ga$_{1-x}$As coupled quantum wells at low temperatures $T \geq 50$ mK and high magnetic fields $B \leq 16$ T. At high magnetic fields cusps are observed in the energy and intensity of the indirect (interwell) exciton photoluminescence. We tentatively attribute these to the commensurability effects of the magnetoexciton with island structures in the sample. The indirect exciton lifetime is found to increase with magnetic field. The increase is attributed to the reduction of indirect exciton localization area caused by the increase of the magnetoexciton mass. The indirect exciton photoluminescence energy is found to enhance with density, which reflects the net repulsive interaction between indirect excitons. [S0163-1829(99)06635-7]

I. INTRODUCTION

In the neutral ideal system of spatially separated electron (e) and hole (h) layers at high perpendicular magnetic fields at low temperatures, a variety of ground states were theoretically predicted, dependent on the relation between the interparticle and interlayer spacings and the magnetic length $l_B = \sqrt{\hbar c/eB}$. For small interlayer separations $d \lesssim l_B$ the ground state is expected to be the exciton condensate as determined by interlayer e-h interaction, whereas for $d \gtrsim l_B$ the ground state is controlled by intralayer e-e and h-h interactions and is expected to be either coupled electron and hole incompressible quantum liquids or Wigner solids.[1–3] The spatially separated e-h system with photoexcitation-controlled e-h density is realized in coupled quantum wells (CQWs) and is remarkable due to the fact that because of much longer e-h recombination time compared to single-layer e-h systems one can reach lower e-h temperatures that are close to the lattice temperature.[4–7]

The expected collective states can be probed by means of photoluminescence (PL) measurements. Studies of the indirect (interwell) exciton PL in AlAs/GaAs CQWs at low temperatures and high magnetic fields showed a large increase in both the exciton diffusivity and the exciton radiative decay rate[8] as well as a huge noise in the integrated exciton PL intensity,[9] which was interpreted as evidence for the exciton condensation. The electron (hole) condensation of a single-layer two-dimensional electron (hole) gas into an incompressible quantum liquid was found to be accompanied by variations of the PL line position and intensity, tracing fractional filling factors, and the formation of a localized state at low filling factors was found to lead to the appearance of new emission lines with long decay time.[10–16]

Here, we perform the detailed investigation of spatially separated electron and hole layers in high-quality GaAs/Al$_x$Ga$_{1-x}$As coupled quantum wells at low temperatures $T \geq 50$ mK and high magnetic fields $B \leq 16$ T using cw and time-resolved PL measurements. Contrary to AlAs/GaAs CQWs of Refs. 8 and 9, the interlayer distance in the studied CQW is relatively large, $d \gtrsim l_B$ at $B > 4$ T, and thus no exciton condensate is expected at high magnetic fields. The behaviors of the PL energy, intensity, and kinetics are analyzed with changing a normal magnetic and electric field as well as e-h density.

II. SAMPLES AND EXPERIMENTAL TECHNIQUE

The electric-field-tunable n^+-i-n^+ GaAs/Al$_x$Ga$_{1-x}$As CQW structure was grown by molecular-beam epitaxy. A sketch of the band diagram of the structure is shown in the top inset to Fig. 1. The i region consists of two 8-nm GaAs QWs separated by a 4-nm Al$_{0.33}$Ga$_{0.67}$As barrier and surrounded by two 200-nm Al$_{0.33}$Ga$_{0.67}$As barrier layers. The n^+ layers are Si-doped GaAs with $N_{Si} = 5 \times 10^{17}$ cm^{-3}. The electric field in the z direction is monitored by the external gate voltage V_g applied between n^+ layers. The small disorder in the CQW is indicated by the indirect exciton PL linewidth of about 1 meV.

Carriers were photoexcited by either a HeNe laser ($\hbar\omega = 1.96$ eV) or a semiconductor laser ($\hbar\omega = 1.95$ eV) operated in the cw or pulsed regime (the pulse duration was about 50 ns, the edge sharpness including the system resolution was ≈ 1 ns, and the repetition frequency was 1 MHz). Exci-
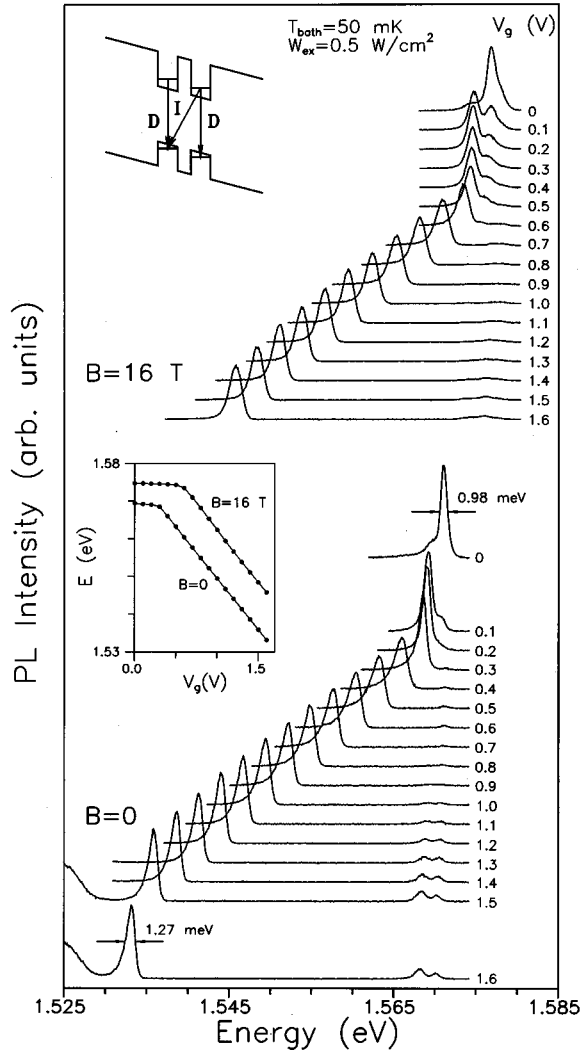
FIG. 1. Gate voltage dependence of the PL spectrum at T_{bath} = 50 mK, W_{ex}=0.5 W/cm^2, B=0, and 16 T. Top inset: schematic band diagram of the GaAs/Al$_x$Ga$_{1-x}$As CQW structure under applied gate voltage; the direct (D) and indirect (I) exciton transitions are shown by arrows. Bottom inset: the ground-state PL line energy as a function of gate voltage.

tation and detection of the PL signal were performed by means of an optical fiber with diameter 0.2 or 0.6 mm positioned 0.3 to 0.6 mm above the mesa. In order to minimize the effect of the mesa heating, the net sample area of about 4 mm^2 was much larger than the mesa area of 350×350 μm^2, and the bottom of the sample was soldered to a metal plate; for cw measurements the excitation was modulated with the dark-to-light ratio of about 15. The measurements were performed in a He^3/He^4 dilution refrigerator with a base temperature T_{bath}=50 mK. The PL kinetics was measured using a time-correlated photon-counting system, while in the cw experiments a charge-coupled device camera or lock-in amplifier was utilized.

III. INDIRECT EXCITON AT A MAGNETIC FIELD

A. Direct-to-indirect crossover

A typical gate voltage dependence of the PL spectrum at low excitation densities is displayed in Fig. 1 for B=0 and

B=16 T; the corresponding ground-state PL line positions are shown in the bottom inset to Fig. 1. For positive V_g <0.3 V at B=0 and V_g<0.6 V at B=16 T, the lowest-energy PL line position does not practically depend on V_g and has short PL decay time (below the system resolution of 1 ns) and, therefore, reflects the direct exciton (so-called direct regime); for higher gate voltages, the main PL line shifts linearly with increasing V_g and has long PL decay time and thus corresponds to the indirect exciton that is constructed from e and h in different layers (the indirect regime). The ratio between the direct and indirect exciton densities in the indirect regime, which is proportional to the ratio between the direct and indirect PL line intensities multiplied by the ratio between the direct and indirect radiative decay times, is vanishingly small. The double structure of the direct exciton line at B=0 is likely to result from the difference in the widths of two QWs. The onset of the broad PL line corresponding to the bulk n^+-GaAs emission is seen at the lowest energies in Fig. 1; this line spreads down to about 1.48 eV and is insensitive to V_g. We have checked that all of the spectra do not practically change upon reversal of V_g polarity.

The crossover between the direct-to-indirect ground state proceeds from the gate voltage behavior of the direct $\mathcal{E}_D$ = $E_g - E_D$ and indirect $\mathcal{E}_I = E_g - E_I - eFd$ exciton energies, where E_g is the energy gap including the electron and hole confinement energies in the CQW, E_D and E_I are the direct and indirect exciton binding energies, e is an electron charge, d is the separation between e and h layers, $F = V_g/d_0$ is the electric field in the z direction, and d_0 is the i-layer width. The $\mathcal{E}_I$ shift equal to eFd allows the determination of the interlayer separation d=11.5 nm, which is very close to the distance between the QW centers. The direct-to-indirect crossover field, $F_{D\text{-}I}$, given by $eF_{D\text{-}I}d = E_D - E_I$, is found to increase with magnetic field (bottom inset to Fig. 1). This corresponds to the stronger enhancement with magnetic field of the direct exciton binding energy compared to E_I;[17,18] particularly, in the high magnetic field limit these energies are evaluated as $E_D \sim 1/l_B$ and $E_I \sim 1/(l_B^2 + d^2)^{1/2}$.

The detailed magnetic-field dependence of the PL spectrum in the indirect regime is presented in Fig. 2. The indirect line $\mathcal{E}_I = E_g + \hbar\omega_c/2 - E_I - eFd$ shifts with B to higher energies stronger than the direct line $\mathcal{E}_D = E_g + \hbar\omega_c/2 - E_D$ (here $\hbar\omega_c$ is the sum of the electron and hole cyclotron energies), which reflects again the stronger enhancement of E_D with magnetic field.

B. Cusps in the energy and intensity of indirect exciton PL

We analyze both the integrated indirect exciton PL intensity $M_0 = \int I(E)dE$ and the PL line position given by the line gravity center $M_1 = M_0^{-1}\int EI(E)dE$ as a function of magnetic field. The excitation energy ($\hbar\omega$=1.96 eV) is chosen to be above the barrier energy to avoid absorption modulation. In principle, the excitation above the barrier may produce deviations of the electron-hole system in the CQW from the charge neutrality because of different collections by the CQW of photoexcited electrons and holes in the barrier layers.[19,20] In our experiments no such deviations are detected. Figure 3 displays the dependences $M_0(B)$ and $M_1(B)$ at different excitation densities (a1)-(b1)-(c1), gate voltages
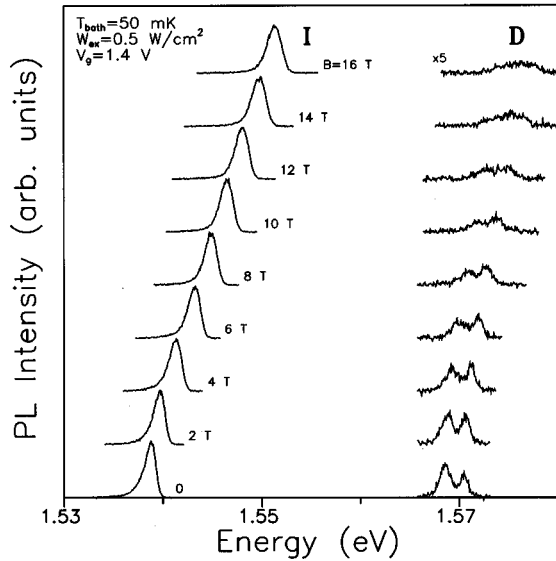
FIG. 2. Magnetic-field dependence of the PL spectrum in the indirect regime ($V_g = 1.4$ V) at $T_{bath} = 50$ mK and $W_{ex} = 0.5$ W/cm^2.

(a2)-(b2)-(c2), and temperatures (a3)-(b3)-(c3). The energy shift of the indirect exciton PL line with magnetic field reflects the excitonic recombination: a quadratic shift at low magnetic fields changes to an approximately linear shift at high fields (Fig. 3).[21,22] The magnitude of the linear shift 0.8 meV/T corresponds to the zero Landau level energy $\hbar eB/2mc$ with $m = 0.072 m_0$, which is higher compared to the reduced electron-hole mass because of the magnetic-field dependence of E_I discussed above. For visualization purposes this shift is subtracted from the dependences $M_1(B)$ to

obtain $\Delta(B)$. As seen from Fig. 3(b), pronounced well-reproducible cusps in energy occur at high fields $B \gtrsim 7$ T. Also, the integrated indirect exciton PL intensity exhibits a similar oscillating behavior that is correlated to a certain extent with the energy variations [Fig. 3(c)]: some of the energy maxima coincide with the maxima of M_0 (dashed lines in Fig. 3), whereas the others coincide with the minima of M_0 (dotted lines in Fig. 3). As is evident from Fig. 3, the cusp position is insensitive to both excitation density and gate voltage, and their amplitude drops with temperature. Within experimental accuracy, no cusps are observed either in the direct regime [Fig. 3(b2)] or when using the semiconductor laser excitation with a bit lower energy $\hbar \omega = 1.95$ eV. The absence of cusps in energy in the direct regime can be related to high effective e-h temperatures (Sec. I); the cusp behavior with excitation energy will be the subject of future studies.

Since the position of the cusps in the energy and intensity of indirect exciton PL is independent of the excitation-controlled exciton density [Fig. 3(b1)], the cusps are not related to either filling-factor-sensitive collective states or magnetic-field-dependent screening and are likely to be of one-magnetoexciton origin. At the same time, no cusps in energy and intensity are expected for both the indirect and direct one-magnetoexciton recombination in ideal single- or double-layer e-h systems.[18,21–23] Therefore, we tentatively attribute the observed cusps to the commensurability effects of the magnetoexciton with island structures in the sample. The cusp presence may reveal the potential correlations in the CQW.

C. Indirect exciton PL kinetics

The indirect exciton PL kinetics in the spatially separated e-h system in CQWs was studied using the same samples at
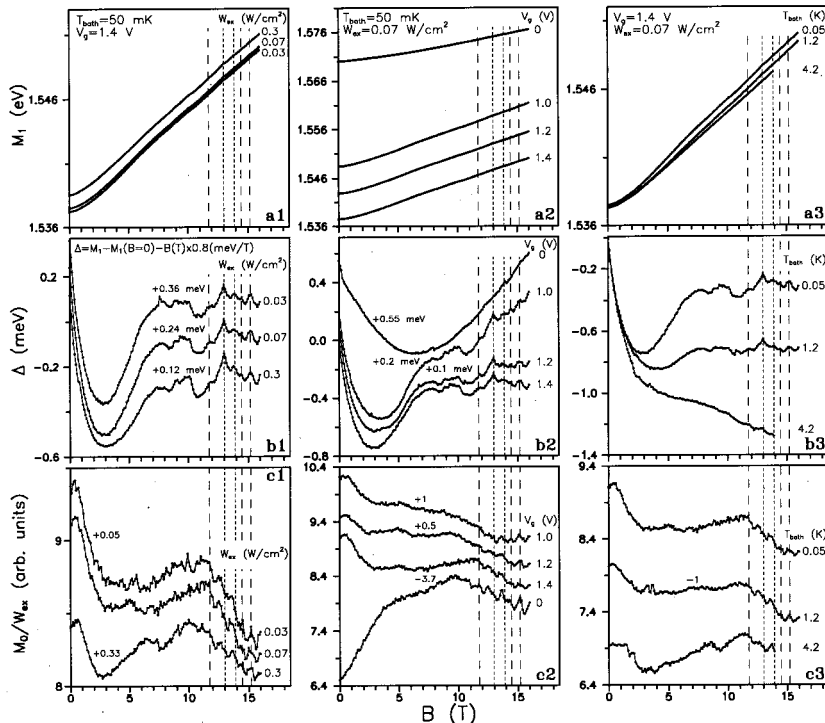

FIG. 3. Magnetic-field dependences of the indirect exciton PL line position M_1 (a), its deviation from the linear shift $\Delta = M_1 - M_1(B=0) - B(T) \times 0.8$ (meV/T) (b), and the integrated indirect exciton PL intensity M_0 normalized by the excitation density (c) vs excitation density (1), gate voltage (2), and temperature (3). In case (2), the data at $V_g = 0$ correspond to the direct exciton for which the slope of the linear shift subtracted in (b2) is equal to 0.4 meV/T. Some of the curves are shifted vertically for clarity; the shift magnitudes are indicated.
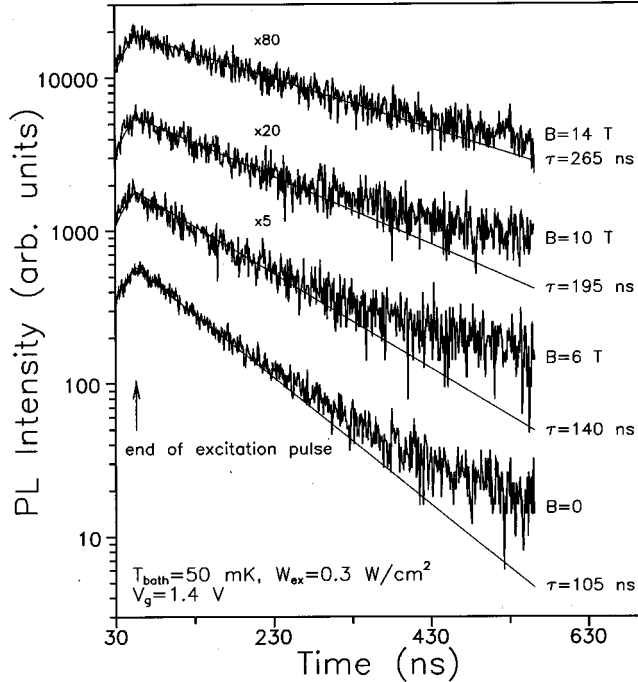
FIG. 4. Magnetic-field dependence of the indirect exciton PL kinetics at $T_{bath}=50$ mK, $W_{ex}=0.3$ W/cm^2, and $V_g=1.4$ V. The thin lines are fitting curves for the initial times of the PL decay. The decay times are indicated.

FIG. 5. PL spectra in the indirect regime ($V_g=1.4$ V) for $W_{ex}=0.5$ and 4 W/cm^2 at $B=0$, 8, and 16 T.

$B=0$: at low temperatures and high excitation densities, an abrupt increase of the spectrally integrated indirect exciton PL intensity was observed right after switching off the excitation[24] and explained in terms of a jump of the population of the optically active small-momentum exciton states.[25–29] The effect is absent at low excitation densities due to the strong localization of excitons in deep potential minima with the localization length smaller than the wavelength of the emitted light,[24] which gives rise to washing out the border between the optically active and inactive states.[30]

Figure 4 represents the magnetic-field dependence of the PL kinetics. The range of excitation densities used corresponds to the low-density regime[24] in which a sharp increase of the indirect exciton PL intensity right after switching off the excitation is not observed. The indirect exciton has a long PL decay time characteristic of the recombination of spatially separated electrons and holes so that the electron-hole system can thermalize down to low effective temperatures.[31] As seen from Fig. 4, the indirect exciton PL decay time increases with magnetic field. We have checked that with varying V_g the integrated indirect PL intensity remains nearly constant while the decay time varies by over three orders of magnitude.[24] Hence, the radiative recombination is dominant in the CQW studied and the observed behavior of the PL decay reflects the increase of the radiative decay time with magnetic field. We note that this is qualitatively different from the case of AlAs/GaAs CQWs where the indirect exciton radiative and nonradiative decay times were observed to drop abruptly at low temperatures and high magnetic fields, which was related to the exciton condensation.[8] In the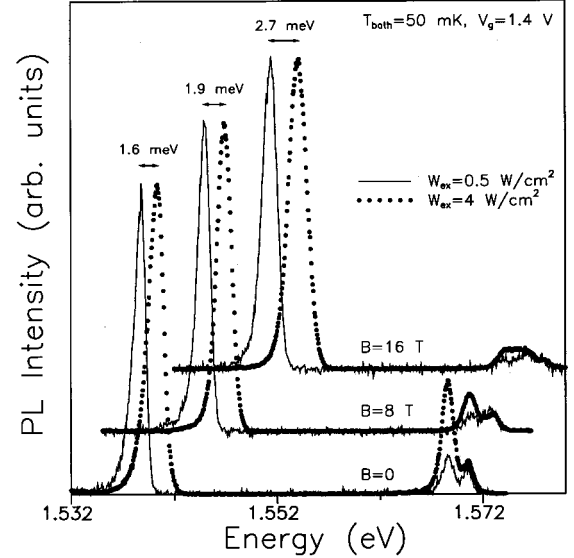 studied GaAs/Al$_x$Ga$_{1-x}$As CQW, where no exciton condensate is expected at high magnetic fields (Sec. I), no such drop of the decay time is observed.

The exciton radiative decay rate is proportional to the dipole matrix element connecting Bloch states in the valence and conduction bands, the overlap between the electron and hole wave functions describing electron-hole relative motion, the lateral size of the exciton center-of-mass wave function (so-called exciton coherent area as determined by the exciton localization length and scattering length), and the occupation of the optically active exciton states.[25–29] Its variation with magnetic field is determined by the competition between the shrinkage of the in-plane exciton radius (given by l_B at high magnetic fields[23]) and the reduction of the exciton localization area, which lead, respectively, to increasing and decreasing the exciton oscillator strength. The observed enhancement of the indirect exciton radiative decay time with magnetic field (Fig. 4) can be explained by the reduction of the exciton localization area at the magnetic field, caused by the magnetoexciton mass increase as calculated in Ref. 23.

D. Indirect exciton PL energy shift with increasing *e-h* density

An example of the PL spectra at two different excitation densities for a number of magnetic fields is shown in Fig. 5. One can see from the figure that, unlike the direct exciton, the indirect exciton energy increases with excitation density at all magnetic fields. This observation is consistent with the theoretically predicted enhancement of the indirect exciton energy with *e-h* density: it can be understood in terms of the net repulsive interaction between indirect excitons caused by the dipole-dipole repulsion for low exciton densities, and in terms of the energy shift originating from the electric field between the separated electron and hole layers for high *e-h* densities[2,35] (in the latter case the energy shift can be roughly estimated using the plate capacitor formula). For fixed excitation density the magnitude of the shift is increased with magnetic field (Fig. 5) mainly due to the increase of the

decay time (and, hence, of the e-h density) with magnetic field (Fig. 4). Note that for fixed exciton density the dipole-dipole repulsion between excitons should be weakened with magnetic field due to the shrinkage of the in-plane exciton radius;[36] this partly suppresses the increase of the shift with magnetic field observed for fixed excitation density (Fig. 5).

Using the plate capacitor formula for determining the energy shift $\delta E = 4\pi n_{eh} e^2 d/\varepsilon$ (where ε is the dielectric constant and n_{eh} is the e-h density), we estimate the exciton density from the data of Fig. 5 at $\approx 9 \times 10^9$ cm^{-2} at $B = 0$ and $\approx 1.5 \times 10^{10}$ cm^{-2} at $B = 16$ T for $W_{ex} = 4$ W/cm^2. As long as the exciton density increases sublinearly with excitation density owing to the reducing decay time,[24] one can just very crudely evaluate the exciton density in the low-density regime (considered in sections A–C), ignoring the decay time variation; e.g., for $W_{ex} = 0.3$ W/cm^2 the density obtained by a linear extrapolation is $\sim 10^9$ cm^{-2}.

The above consideration of the energy shift is relevant for the case of delocalized excitons for which this shift is determined by exciton-exciton interaction. For the case of localized excitons in a random potential, the enhancement of the indirect exciton PL energy with e-h density follows (apart from the interaction) from the finite degeneracy of the 0D exciton state. Disregarding the exciton-exciton interaction, the degeneracy can be estimated at $\sim S/a_B^2$, where S is the exciton localization area and a_B is the exciton Bohr radius. Due to the finite degeneracy, the lower-energy localized exciton states are successively filled with increasing exciton density, which results in the enhancement of the mean exciton energy. Apparently, the finite degeneracy effect should be dominant at low exciton densities while for high densities the energy shift should mainly be determined by the interaction effect discussed above.

IV. CONCLUSIONS

The photoluminescence of the spatially separated electron and hole layers in GaAs/Al$_x$Ga$_{1-x}$As coupled quantum wells has been studied at low temperatures $T \geqslant 50$ mK and high magnetic fields $B \leqslant 16$ T. The electric field at which the direct-to-indirect ground-state crossover occurs is found to increase with magnetic field due to the stronger enhancement of the direct exciton binding energy compared to that of indirect exciton. At high magnetic fields, cusps are observed in the energy and intensity of the indirect exciton photoluminescence. The obtained excitation density, gate voltage, and temperature dependences show that the cusps are of one-exciton origin. We tentatively attribute them to the commensurability effects of the magnetoexciton with island structures in the sample. The indirect exciton lifetime is found to increase with magnetic field. The increase is attributed to the reduction of indirect exciton localization area caused by the increase of the magnetoexciton mass. The indirect exciton photoluminescence energy is found to enhance with density, which reflects the net repulsive interaction between indirect excitons.

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