From spatially indirect excitons to momentum-space indirect excitons by an in-plane magnetic field

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An in-plane magnetic field is found to change drastically the photoluminescence spectra and kinetics of interwell excitons in GaAs/$Al_xGa_{1-x}As$ coupled quantum wells. This effect is due to the in-plane magnetic-field-induced displacement of the interwell exciton dispersion in momentum space, which results in the transition from the momentum-space direct exciton ground state to the momentum-space indirect exciton ground state. An in-plane magnetic field is, therefore, an effective tool for exciton dispersion engineering.

The effective exciton temperature in a quasiequilibrium system of excitons in semiconductors is determined by the ratio between the exciton energy relaxation rate and the exciton recombination rate. The low exciton temperature is crucial for an observation of a number of novel collective phenomena caused by the high occupation of the lowest-energy exciton states in a quasi-two-dimensional (2D) exciton system.[1–5] Low temperatures can be achieved in a system with a low exciton recombination rate.

A long exciton lifetime is characteristic (i) for systems where the ground-state exciton is optically inactive (in the dipole approximation) because of parity, e.g., for Cu_2O; (ii) for systems where the electron and hole are spatially separated, e.g., for spatially indirect (interwell) excitons in direct-band-gap coupled quantum wells (CQW's) such as $\Gamma-X_z$ AlAs/GaAs CQW's and GaAs/$Al_xGa_{1-x}As$ CQW's; (iii) for systems where the ground-state exciton is indirect in momentum space, e.g., for $\Gamma-X_{xy}$ AlAs/GaAs CQW's.

Due to the coupling between the internal structure of a magnetoexciton and its center-of-mass motion,[6] the ground exciton state in a direct-band-gap semiconductor in crossed electric and magnetic fields was predicted to be at finite momentum. In particular, the transition from the momentum-space direct exciton ground state to the momentum-space indirect exciton ground state was predicted (i) for an interwell exciton in coupled quantum wells in an in-plane magnetic field,[1,7] and (ii) for a single-layer exciton in an in-plane electric field and perpendicular magnetic field.[8–10] These effects should allow for a controllable variation of the exciton dispersion and an increase of the exciton ground-state lifetime.

In the present paper, we report on the experimental observation of the in-plane magnetic-field-induced transition from the momentum-space direct exciton ground state to the momentum-space indirect exciton ground state for a system of interwell excitons in GaAs/$Al_xGa_{1-x}As$ CQW's. The transition is identified by the drastic change of the exciton photoluminescence (PL) kinetics. The interwell exciton dispersion in an in-plane magnetic field is analyzed theoretically and determined experimentally from the shift of the interwell exciton PL energy; the experimental data are found to be in qualitative agreement with the theory.

We suppose that the in-plane magnetic field is directed along the x axis, and we use the calibration $\mathbf{A}=\mathbf{e}_y Bz$. In the calibration used, the center-of-mass in-plane momentum $\mathbf{P}_\parallel(P_x,P_y)$ is the integral of motion. The effective Schrödinger equation for an exciton can be presented in the form $(\hat{\mathbf{H}}_0+\hat{\mathbf{H}}_z)\Psi=E\Psi$, where

$$\hat{\mathbf{H}}_0=\frac{P_x^2+\left(P_y-\frac{e}{c}Bd\right)^2}{2M}, \tag{1}$$

and $\hat{\mathbf{H}}_z$ is the operator depending only on z_e and z_h counted from the centers of the corresponding quantum wells; d is the mean separation between the electron and hole layers. The value $p_B=-(e/c)Bd=\hbar d/l_B^2$ is the shift of the magnetic momentum of an interwell exciton in the ground state (as follows from the analysis of the Schrödinger equation); $l_B=(eB/\hbar c)^{-1/2}$ is the magnetic length.

The physical sense of this shift can be obtained from an analysis of the adiabatic turning on of the in-plane magnetic field. The appearance of the vortex electric field leads to the acceleration of an interwell exciton. The final in-plane momentum is equal precisely to $-(e/c)Bd$ and directed normal to the magnetic field. Therefore, the appearance of the momentum $-(e/c)Bd$ is related to the diamagnetic response of the electron-hole (exciton) system in CQW in an in-plane magnetic field.

The contribution of the second order on the magnetic field consists of two parts: the first, depending on the momenta, is the sum of Van Vleck paramagnetism of isolated QW's, and the second is the sum of diamagnetic shifts of isolated QW's.[11] Van Vleck paramagnetism leads to the renormalization of the effective (magnetic) mass of an exciton along the y axis, M_{yy}. Therefore, the magnetoexciton dispersion law becomes anisotropic,

$$M_{xx}=M=m_e+m_h, \quad M_{yy}=M+\delta M(B), \tag{2}$$

where

$$\delta M(B) = \frac{e^2 B^2}{c^2}(f_e + f_h), \tag{3}$$

$$-f_{e,h} = \sum_n \frac{|\langle 0|z_{e,h}|n\rangle|^2}{E_0 - E_n}. \tag{4}$$

Here $E_n, |n\rangle$ are the energies and state vectors corresponding to the size quantization of QW's; $f_{e,h}$ are related to the electric polarization of QW's in the z direction. $\delta M(B) > 0$. The estimate results to

$$\frac{\delta M_{yy}}{M} \sim \frac{E_{\text{diam}}}{E} \sim \left(\frac{L_z}{l_B}\right)^4, \tag{5}$$

where E_{diam} is the diamagnetic shift in the isolated QW, E is the size-quantized excitation energy of the QW, and L_z is the QW width.

The electric-field-tunable n^+-i-n^+ GaAs/Al_xGa_{1-x}As CQW structure was grown by molecular-beam epitaxy. The i region consists of two 8 nm GaAs QW's 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 $N_{\text{Si}} = 5\times10^{17}$ cm^{-3}. The electric field in the z direction is monitored by the external gate voltage V_g applied between n^+ layers. Carriers were photoexcited by a pulsed semiconductor laser ($\hbar\omega = 1.85$ eV, the pulse duration was about 50 ns, the edge sharpness including the system resolution was ≈ 0.5 ns, the repetition frequency was 1 MHz, and $W_{\text{ex}} = 10$ W/cm^2). In order to minimize the effect of the mesa heating, we worked with a mesa area of 0.2×0.2 mm^2, which was much smaller than the sample area of about 4 mm^2. In addition, the bottom of the sample was soldered to a metal plate. The PL measurements were performed in a He cryostat by means of an optical fiber with diameter 0.1 mm positioned 0.3 mm above the mesa. The PL spectra and kinetics were measured by a time-correlated photon-counting system.

The separation of electrons and holes in different QW's (the indirect regime) is realized by applying a finite gate voltage which fixes an external electric field in the z direction $F = V_g/d_0$, where d_0 is the i-layer width.[12] The spatially direct (intrawell) and indirect (interwell) transitions are identified by the PL kinetics and gate-voltage dependence: the intrawell PL line has a short PL decay time and its position practically does not depend on V_g, while the interwell PL line has a long PL decay time and shifts to lower energies with increasing V_g (the shift magnitude is given by eFd).[12,13] The upper and lower direct transitions are related to the intrawell $1s$ heavy-hole exciton X and the intrawell charged complexes X^+ and X^-.[14]

With increasing in-plane magnetic field, the energy of the interwell exciton increases while the energies of the direct transitions are almost unaffected [Fig. 1(a)]. This behavior is consistent with a displacement of the interwell exciton dispersion in an in-plane magnetic field. The scheme of the interwell exciton dispersion at zero and finite in-plane magnetic field is shown in Fig. 2.

For delocalized 2D excitons, only the states with small center-of-mass momenta $k \leqslant k_0 \approx E_g\sqrt{\varepsilon_b}/\hbar c$ (where ε_b is the background dielectric constant), i.e., within the radiative zone, can decay radiatively[15] (see Fig. 2). For GaAs structures, $k_0 \approx 3\times10^5$ cm^{-1} and is much smaller than $k_B = p_B/\hbar$ in strong fields (at $B = 10$ T, $k_B \approx 2\times10^6$ cm^{-1}). The energy of the interwell exciton PL is set by the energy of the radiative zone. Therefore, as the diamagnetic shift of the bottom of the subbands is small and can be neglected to the first approximation, the interwell exciton PL energy in an in-plane magnetic field should be increased by $E_{p=0} = p_B^2/2M = e^2d^2B^2/2Mc^2$. In particular, the in-plane magnetic-field dependence of the interwell PL energy could be used for the measurement of the exciton dispersion because it allows us to access high momentum exciton states. At small fields, the measured PL energy shift rate is 0.062 meV/T^2 [see the inset to Fig. 1(a)], which corresponds to $M = 0.21m_0$. This value is close to the calculated mass of the heavy-hole exciton in GaAs QW's $\approx 0.25m_0$ ($m_e = 0.067m_0$ and the calculated in-plane heavy-hole mass near $k = 0$ is reported to be $m_h = 0.18\ m_0$, see Ref. 16 and references therein). Figure 1 shows a considerable deviation from the quadratic dependence of the interwell exciton PL energy at high B. This deviation is a consequence of the interwell exciton mass renormalization due to an in-plane magnetic field, which is predicted by theory, see Eqs. (2)–(4). Due to the estimate of Eq. (5), the deviation of the in-

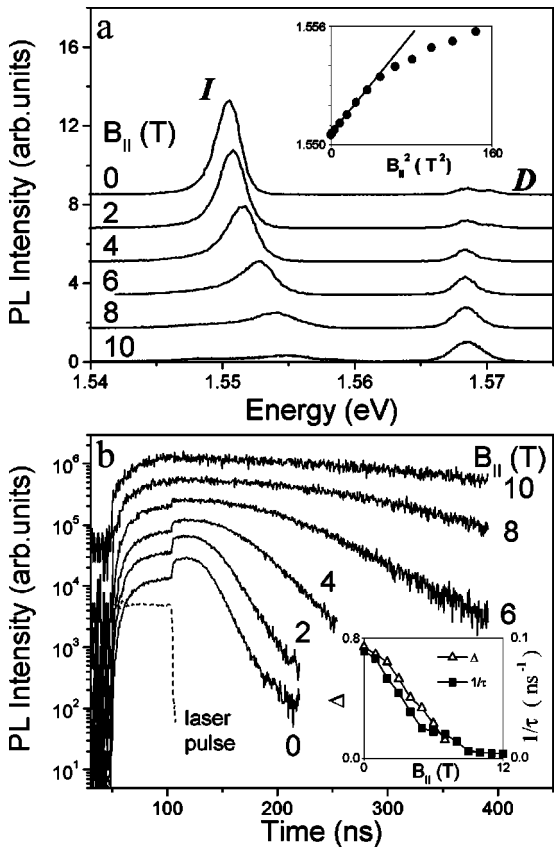

FIG. 1. In-plane magnetic field dependence of the time-integrated PL spectrum (a) and the interwell exciton PL kinetics measured at the maximum of the interwell exciton spectrum (b) in the indirect regime ($V_g = 1$ V) at $T_{\text{bath}} = 1.5$ K. Upper inset: The interwell exciton energy vs in-plane magnetic field; the line is the fitting curve for small fields $E_{p=0} = e^2d^2B^2/2Mc^2$, see text. Lower inset: The fastest interwell exciton PL decay rate and the interwell exciton PL intensity enhancement after excitation switch off, $\Delta = \ln(I_{\text{PL-max}}/I_{\text{PL-pulse end}})$, vs in-plane magnetic field.

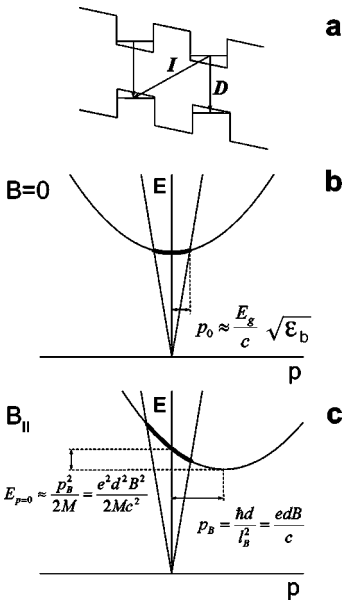

FIG. 2. Schematic band diagram of the GaAs/$Al_xGa_{1-x}As$ CQW (a). Scheme of the interwell exciton dispersion at zero (b) and finite (c) in-plane magnetic field; the photon dispersion as well as the radiative zone marked bold are also shown.

terwell exciton dispersion from the quadratic one should become essential in the magnetic field where $l_B \sim L_z$. This is in good qualitative agreement with the experiment which presents the onset of the deviation at $B \approx 8$ T, where $l_B = 9$ nm$\sim L_z = 8$ nm [see the inset to Fig. 1(a)]. A possible contribution from the hole dispersion nonparabolicity to the observed increase of the exciton mass is, apparently, a minor effect for the small exciton energies considered, which are much smaller than the light-hole–heavy-hole splitting equal to 17 meV for the CQW studied.[14]

Note that the interwell exciton dispersion in an in-plane magnetic field is related to the single-particle dispersions. For separated electron layers, an in-plane magnetic field results in the relative displacement of the electron dispersion parabolas, which has been observed in resonant tunneling[17–20] and in-plane transport[21–24] experiments. In particular, the development of an indirect gap when an in-plane magnetic field is applied has been observed in an asymmetric modulation-doped single quantum well where the centers of the electron and hole envelopes do not coincide; due to the free-carrier character of recombination in the studied 2D electron gas, the PL energy shift corresponded to the electron mass.[25]

An in-plane magnetic field modifies qualitatively the exciton PL kinetics [Fig. 1(b)]. The main feature of the interwell exciton PL kinetics at zero magnetic field is a sharp enhancement of the PL intensity after the excitation is switched off—the PL jump[13] (Fig. 1). The basis of the effect is the following. The exciton PL kinetics is determined by the kinetics of the occupation of the radiative zone (marked bold in Fig. 2). The occupation varies due to the exciton recombination and energy relaxation. The PL jump denotes a sharp increase of the occupation of the optically active exciton states just after the excitation is switched off. It is induced by the sharp reduction of the effective exciton temperature, T_{eff}, due to the fast decay of the nonequilibrium

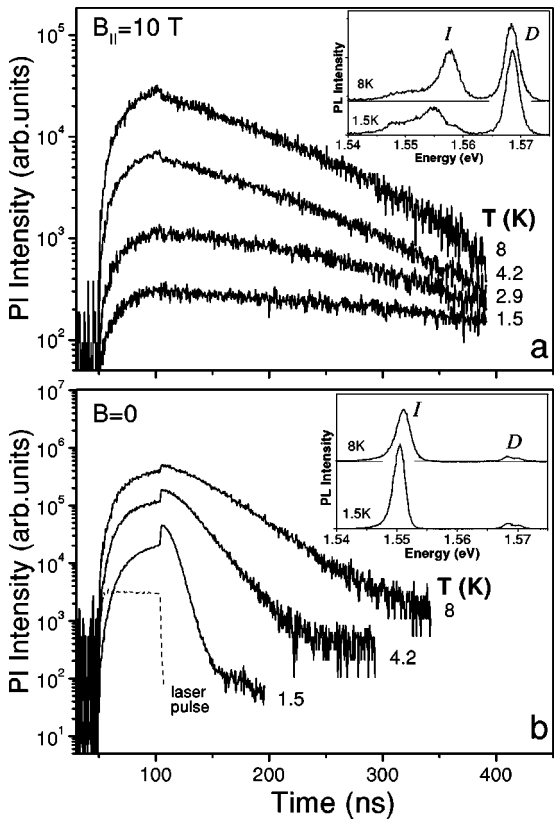

FIG. 3. The interwell exciton PL kinetics measured at the maximum of the interwell exciton spectrum vs temperature in an in-plane magnetic field $B = 10$ T (a) and at $B = 0$ (b). Insets: the corresponding time-integrated PL spectra.

phonon occupation and energy relaxation of hot photoexcited excitons, electrons, and holes.[13] The disappearance of the PL jump at high in-plane magnetic fields is consistent with the displacement of the interwell exciton dispersion in a parallel magnetic field: for a momentum-space indirect exciton, a sharp reduction of T_{eff} just after the excitation is switched off should not result in an increase of the occupation of the radiative zone [see Fig. 2(c)] and, hence, the PL intensity.

The measured radiative decay rate is proportional to the fraction of excitons in the radiative zone. The observed strong reduction of the radiative decay rate in an in-plane magnetic field [by more than 20 times in $B = 12$ T, see Fig. 1(b)] also reflects the displacement of the interwell exciton dispersion and, correspondingly, the nonradiative character of the ground exciton state in a parallel magnetic field. In a high in-plane magnetic field, the radiative decay rate becomes comparable to and smaller than the nonradiative decay rate which results in the observed quenching of the interwell exciton PL intensity [Fig. 1(a)].

The unambiguous evidence for the nonradiative character of the ground state of an interwell exciton in a parallel magnetic field has been observed from the temperature dependence of PL kinetics (Fig. 3). At zero field, the exciton recombination rate monotonically *reduces* with increasing temperature [Fig. 3(b)] due to the thermal reduction of the radiative zone occupation.[13] In a high in-plane magnetic field, the temperature dependence is the opposite: the exciton recombination rate *enhances* with increasing temperature [Fig. 3(a)] due to the increasing occupation of the radiative

zone. Correspondingly, with increasing temperature the interwell exciton PL intensity reduces at zero field and enhances in a high in-plane magnetic field (see the insets to Fig. 3).

In conclusion, we have observed a drastic change of the photoluminescence spectra and kinetics of interwell excitons in GaAs/$Al_xGa_{1-x}As$ CQW's in an in-plane magnetic field. The effect is due to the in-plane magnetic-field-induced displacement of the indirect exciton dispersion in momentum space, which results in the transition from the momentum-space direct exciton ground state to the momentum-space indirect exciton ground state. An in-plane magnetic field is, therefore, an effective tool for the exciton dispersion engineering and controllable increase of the exciton ground-state lifetime. We speculate that it can be used for the experimental realization of an ultralow-temperature exciton gas, which might result in an observation of predicted collective phenomena caused by the high occupation of the lowest-energy exciton states. In addition, the renormalization of the exciton mass due to an in-plane magnetic field was observed; the experimental data are in qualitative agreement with the theory.

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