Condensation of Indirect Excitons in Coupled AlAs/GaAs Quantum Wells

L. V. Butov,* A. Zrenner, G. Abstreiter, G. Böhm, and G. Weimann
Walter Schottky Institut, Am Coulombwall, D-85748 Garching, Germany
(Received 23 August 1993)

The photoluminescence of excitons confined in an electric field tunable coupled AlAs/GaAs quantum well has been investigated at $T \geq 350$ mK and magnetic fields $H \leq 14$ T. In the indirect regime where electrons and holes are separated both in real and in k space, the magnetic field was found to result in (i) a strong change of both the photoluminescence intensity and decay time which is attributed to anomalies in the exciton transport and (ii) an appearance of a huge broad band noise in the photoluminescence intensity which is evidence for exciton condensation.

PACS numbers: 71.35.+z, 73.20.Dx, 78.20.Ls

The electron-hole (e-h) interaction in the neutral e-h system has been predicted to cause the condensation of bound e-h pairs (excitons) into an excitonic insulator state [1–4]. In the case of a dilute excitonic gas ($na_B^d \ll 1$, a_B—excitonic Bohr radius, n—e-h density, d—dimensionality) the excitons can be considered as rigid Bose particles and the condensation is analogous to the Bose-Einstein condensation of bosons, while in the case of the dense e-h system ($na_B^d \gg 1$) the condensed state is analogous to the BCS superconducting state. The transition between those two limits is smooth [3]. The condensation condition can be achieved only if the temperature of the excitons is below a critical temperature T_c. This criterion is hard to achieve experimentally.

The experimental efforts to observe exciton condensation in bulk semiconductors were concentrated mainly on the analysis of the photoluminescence (PL) line shape and the transport properties of excitons [5–8]. Degenerate Bose-Einstein statistics (which is a precursor of exciton condensation) have been reported for excitons in Cu_2O [5] and in Ge [6] and for biexcitons in CuCl [7]. Rapid exciton transport observed in Cu_2O has been attributed to exciton condensation [8].

In 2D systems the precursor of the exciton condensation, namely the formation of excitons in a dense e-h magnetoplasma, has been observed in InGaAs quantum wells (QWs) [9]. Because of the high rate of carrier recombination the temperature of the photoexcited carriers, however, is higher than T_c. A strong suppression of the recombination rate can be achieved in coupled QWs (CQWs) with spatially separated electrons and holes. CQWs are therefore good candidates for the observation of exciton condensation [10, 11]. Previously reported variations of the PL linewidth [11] in CQWs, however, cannot be interpreted in terms of exciton condensation since the linewidth was found to be dominated by random potential fluctuations [12].

We have studied the PL of a neutral e-h system in an electric field tunable AlAs/GaAs CQW structure (Fig. 1). In this structure electrons and holes are separated both in real and k space, the overlap between them can be controlled by an external gate voltage V_g [13]. The lifetime in this system is sufficiently long to allow for a thermalization of the indirect excitons down to temperatures below 1 K. A perpendicular magnetic field increases the exciton binding energy and suppresses the kinetic energy of the excitons which results in an increase of T_c [14, 15]. Therefore magnetic field has been applied in the present work to improve the critical conditions for exciton condensation. For spatially separated electrons and holes the condensed excitonic phase has been predicted to be the ground state for $s < l_H$ (s is an interlayer separation, l_H is a magnetic length) [16, 17] at $T < T_c$.

Our samples are designed as electric field tunable n^+-i-n^+ structures. They have been grown by molecular beam epitaxy. The active part of the structure is the i-

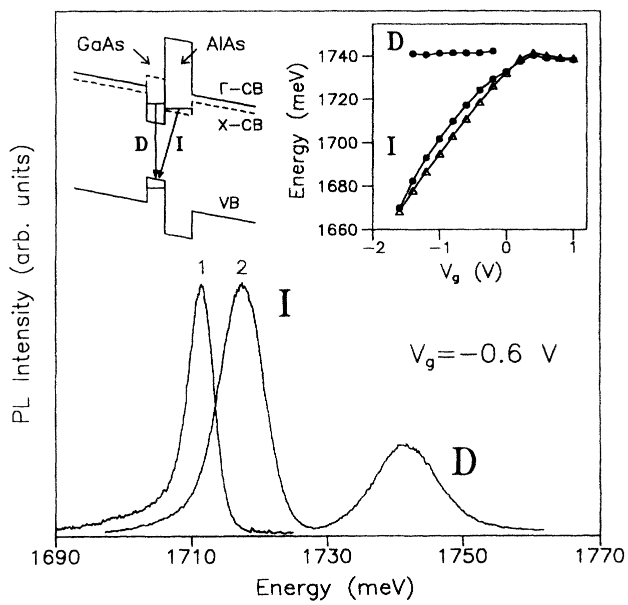

FIG. 1. Normalized PL spectra in the indirect regime ($V_g = -0.6$ V) at $T = 350$ mK for excitation densities of $W_{ex} = 50$ (1) and 500 W/cm^2 (2). Left inset: Band diagram of the real and k space indirect AlAs/GaAs CQW for negative V_g. Right inset: V_g dependence of the direct (D) and indirect (I) transitions at $T = 350$ mK for $W_{ex} = 50$ (triangles) and 500 W/cm^2 (points). For clarity the direct line is only shown for $W_{ex} = 500$ W/cm^2.

0031-9007/94/73(2)/304(4)$06.00

region, which consists of a 40 Å AlAs and 30 Å GaAs layer, surrounded by two 400 Å $Al_{0.48}Ga_{0.52}As$ barrier layers (Fig. 1). Carriers were photoexcited in the GaAs QW by either a cw dye laser ($\hbar\omega = 1.85$ eV) or a pulsed semiconductor laser ($\hbar\omega = 1.8$ eV, $\tau = 4$ ns). Time resolved and cw photoluminescence measurements have been performed at low temperatures ($T \geq 350$ mK) and at high perpendicular ($B_\perp$) and parallel ($B_\parallel$) magnetic fields $B \leq 14$ T.

The direct and indirect excitonic transition energies are shown in the inset of Fig. 1 as a function of V_g for different excitation densities (W_{ex}). In the direct regime ($V_g > 0.4$ V) both electrons and holes are confined in the GaAs QW. In the indirect regime ($V_g < 0.4$ V) electrons are in the AlAs QW and holes in the GaAs QW. Increasing exciton density results in a monotonic increase of the indirect PL energy (E_{PL}). This is caused mainly by the energy shift originating from the electric field between the separated electron and hole layers [16]. As a function of V_g the maximum shift of E_{PL} for fixed W_{ex} (see inset of Fig. 1) corresponds to the maximum exciton density which is determined by the maximum exciton lifetime. Close to the direct regime the exciton lifetime is reduced due to Γ-X mixing [13], while at $V_g < -1$ V it is reduced by tunneling of carriers through the AlGaAs barriers. The monotonic increase of the exciton energy with density is important as it prevents the condensation into metallic e-h droplets [6, 16]. Normalized PL spectra in the indirect regime ($V_g = -0.6$ V) are shown in Fig. 1 for excitation densities $W_{ex} = 50$ (1) and 500 W/cm^2 (2). At 500 W/cm^2 the indirect PL line (I) broadens, indicating the appearance of an e-h plasma.

In the indirect regime the integrated PL intensity (I_{PL}) drops almost 2 orders of magnitude compared to the direct regime. In the indirect regime the radiative lifetime (τ_r) is much larger than the nonradiative lifetime (τ_{nr}) and the total recombination lifetime (τ) is determined by τ_{nr} ($\tau \approx \tau_{nr}$). τ was measured to be 100 ns for $V_g = -0.5$ V, for example.

The application of a magnetic field was found to result in a strong change of both I_{PL} and τ in the indirect regime. This is shown in Fig. 2 for both $B_\perp$ and parallel $B_\parallel$ for $V_g = -0.4$ V and $T = 350$ mK on a logarithmic scale. With increasing $B_\perp$ both I_{PL} and τ first increase and then decrease dramatically. Typical PL spectra at different $B_\perp$ are shown in the inset. $B_\parallel$ results in a monotonic increase of I_{PL} and τ. These effects disappear at temperatures above 5 K where only a small monotonic increase of I_{PL} (about a factor of 2 for $B_\perp = 14$ T) is observed. The variations of I_{PL} and τ are correlated which means that τ_{nr} is subjected to a strong change by magnetic field. Nonradiative Auger recombination is not efficient in our structures since the increase of density does not lead to a reduction of τ. The nonradiative recombination occurs at nonradiative recombination centers (NC). The low temperature diffusivity of excitons in QWs is determined by interface roughness scattering and is very low [18, 19], the rate of nonradiative recombination at the NC is determined by the transport of excitons to the NC [20]. We conclude therefore that magnetic field has a strong influence on the transport properties of excitons. An increase of $B_\perp$ firsts leads to a reduction of the exciton diffusivity and then to a strong increase. Increasing $B_\parallel$ leads to a monotonic reduction of the diffusivity.

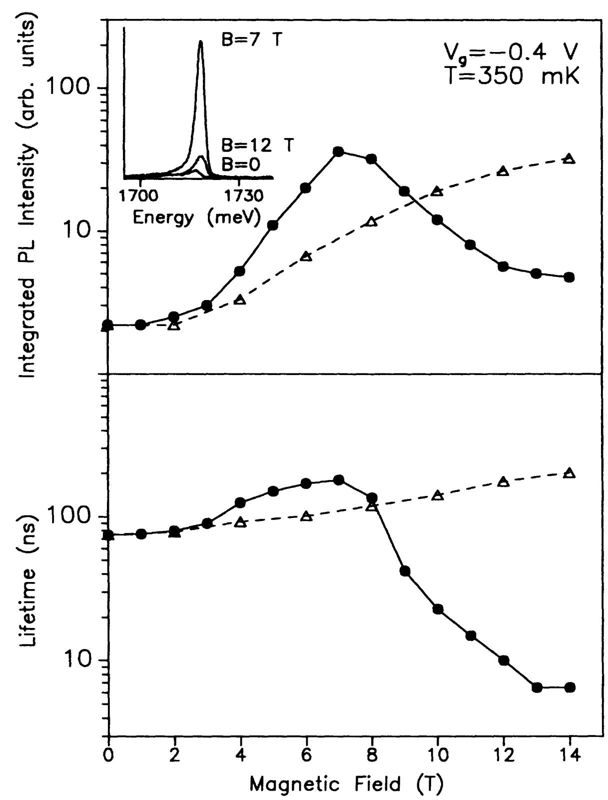

FIG. 2. Integrated PL intensity and decay time in the indirect regime $V_g = -0.4$ V at $T = 350$ mK versus $B_\perp$ (points) and $B_\parallel$ (triangles) in the logarithmic scale. Inset: Corresponding time integrated PL spectra at $V_g = -0.4$ V for $B_\perp = 0, 7$, and 12 T (linear scale).

The influence of the excitonic transport on I_{PL} (huge positive and negative magnetoluminescence) is observed only in the indirect regime where the condition $(D\tau_r)^{1/2} > n_{NC}^{-1/2}$ is valid (D is the diffusion constant and n_{NC} is the NC concentration), which means that excitons can reach the NC before they recombine radiatively. In the indirect regime τ_r is very large and the transport properties of excitons are important.

The observed reduction of the exciton diffusivity in magnetic field can be qualitatively explained by the coupling between the magnetoexciton motion and its internal structure. An exciton moving in a perpendicular field acquires a dipole moment because the Lorentz force acts on electron and hole in opposite directions. This can lead to increased exciton scattering by interface roughness and thus to the observed reduction of the exciton diffusivity.

However, this mechanism is not able to explain the dramatic increase of the exciton diffusivity at high $B_\perp$. This striking anomaly in the magnetoexciton transport seems to be connected with the coherent effects in the exciton system. This is confirmed by the fact that the effect disappears at higher temperatures ($T > 5$ K). The increase of the exciton diffusivity with magnetic field is reminiscent of negative magnetoresistance in electron transport occurring both in the regime of weak [21] and strong [22] localization. The magnetic field changes the phases of the charged particles which results in the suppression of coherent backscattering [21] and destructive interference between the forward paths [22], leading to an increase of the electron mobility (huge negative magnetoresistance is observed in the regime of strong localization [23]). As the exciton is composed of two charged particles, similar effects can be relevant for the exciton transport. However, the direct analogy with electron transport is not straightforward due to the importance of the electron-hole interaction for excitons. The question of how far this mechanism alone can explain the observed huge increase of the exciton diffusivity at high $B_\perp$ has to be answered by future theories of exciton transport in magnetic fields.

A spectacular effect is observed in the indirect regime at perpendicular magnetic fields, namely a huge broad band noise in I_{PL}. The time dependence of the PL signal at $B_\perp = 11$ T and $V_g = -0.5$ V is shown in Fig. 3. For comparison data with about the same PL intensity are shown for $B = 0$ T (achieved with 6 times higher W_{ex}). The observed noise amplitude at $B_\perp = 11$ T is found to be anomalously large as compared to the case of $B = 0$ T, which represents an upper limit for the noise amplitude that can be expected from Poisson statistics for randomly emitted photons originating from single exciton decay. The power spectrum of the anomalously large noise has a broad band spectrum (see inset of Fig. 3).

The appearance of the huge noise is strong evidence for the presence of coherence in the exciton system. The noise amplitude is known to be inversely proportional to the number of statistically independent entities in a system [24]. Large noise amplitudes therefore denote that only a small number of entities exists in the macroscopically large photoexcited region.

We believe that the appearance of those macroscopic entities in the exciton system is given by the condensation of indirect excitons. A condensed domain can be considered as one macroscopic entity. The PL signal of condensed excitons is much higher as compared to uncondensed ones [25]. This is mainly due to the fact that the oscillator strength of the excitonic PL is increased with the increase of the coherent area [26,27] which is given by the domain size for condensed excitons. The formation and disappearance of condensed domains results therefore in a change of the total PL signal. For the existence of broad band noise the dephasing time (during which the phase coherence is maintained within a condensed domain) should have a broad distribution [28].

The following conditions under which the noise is observed confirm this interpretation.

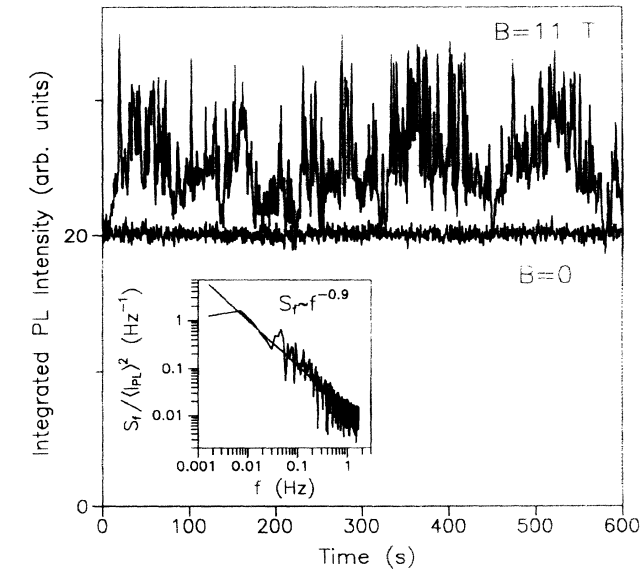

FIG. 3. The time dependence of the PL signal in the indirect regime ($V_g = -0.5$ V) at $T = 350$ mK for $B_\perp = 11$ T ($W_{ex} = 1$ W/cm^2) and at $B = 0$ ($W_{ex} = 6$ W/cm^2). The excitation density for $B = 0$ is higher than for $B_\perp = 11$ T to get approximately the same signal level. Inset: Power spectrum of the noise for $B_\perp = 11$ T.

(i) The noise is observed only in the indirect regime for excitons with long lifetime.

(ii) The noise is observed only at low temperatures. The characteristic temperature dependence of the noise amplitude $\langle \delta I_{PL}\rangle / \langle I_{PL}\rangle$ (see inset of Fig. 4) shows that it is increasing slightly with temperature for $T < 2$ K; between $T = 2$ and 4 K the noise gradually disappears. The decrease of the noise amplitude with increasing temperature reflects a reduction of the average size of condensed domains. The temperature dependence of the transition is smooth which can be explained by a size distribution of the domains. As a function of exciton density the transition temperature first increases and then decreases when approaching plasma densities. It is always in the range below 4 K.

(iii) The noise is observed only in the exciton regime and disappears at high carrier densities corresponding to the plasma regime (the transition to the latter has been determined by the fact that in the plasma regime the PL intensity is saturated and the increase of the excitation power results in a broadening of PL line). A condensation of excitons in the plasma regime is unlikely in the present system due to very different electron and hole Fermi surfaces [1–4].

(iv) The noise is observed only at high $B_\perp$ which improves the condensation conditions. The range of magnetic fields over which the noise is observed is presented in Fig. 4, which shows the dependence of I_{PL} versus $B_\perp$. It can be also seen that the noise is strongly suppressed at the highest fields corresponding to the reduced excitonic lifetime (see Fig. 2). This is in agreement with the expected

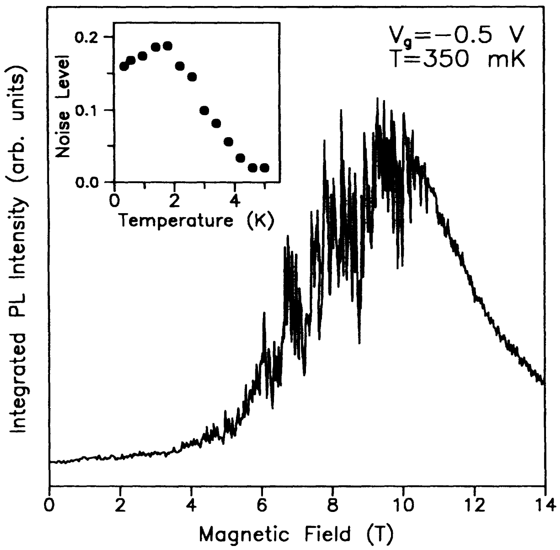

FIG. 4. PL intensity in the indirect regime ($V_g = -0.5$ V) at $T = 350$ mK and ($W_{ex} = -0.5$ W/cm^2) versus $B_\perp$. Inset: Temperature dependence of the noise level $\langle \delta I_{PL} \rangle / \langle I_{PL} \rangle$ in the indirect regime ($V_g = -0.5$ V) at $B = 9$ T.

conditions for the exciton condensation, namely the long lifetime of the excitons. In this range of $B_\perp$ the diffusivity of excitons is high. Therefore we suppose that even if condensed domains are formed they exist only for a short time due to the fast transport of excitons to the NCs. The formation and disappearance of the shortlived domains cannot produce noise in the range of low frequencies which is detectable in our experiment. The possible superfluidity of the condensed excitons can also be responsible for the observed fast exciton transport at high $B_\perp$.

In conclusion, we have observed strong evidence for condensation of indirect excitons confined in coupled AlAs/GaAs QWs which manifests itself as the appearance of a huge broad band noise in the PL intensity. The condensation occurs (i) for indirect excitons with separated electrons and holes, (ii) at low temperatures ($T < 4$ K), (iii) in the excitonic regime, and (iv) at high perpendicular magnetic field. In addition huge negative and positive magnetoluminescence has been observed, which originates from anomalies in the transport of indirect excitons.

We would like to thank G. E. W. Bauer, A. B. Dzyubenko, Al. L. Efros, V. D. Kulakovskii, and A. Odintsov for helpful discussions. L. V. B. thanks FVS foundation for financial support.

---

*Permanent address: Institute of Solid Physics, Russian Academy of Science, 142432 Chernogolovka, Moscow district, Russia.

[1] L. V. Keldysh and Yu. E. Kopaev, Fiz. Tverd. Tel. **6**, 2791 (1964) [Sov. Phys. Solid State **6**, 2219 (1965)].
[2] B. I. Halperin and T. M. Rice, in *Solid State Physics*, edited by H. Ehrenreich and D. Turnbull (Academic, New York, 1968), Vol. 21, p. 115.
[3] C. Comte and P. Nozières, J. Phys. **43**, 1069 (1982); P. Nozières and C. Comte, J. Phys. **43**, 1083 (1982).
[4] G. E. W. Bauer, in *Condensed Systems of Low Dimensionality*, edited by L. J. Beeby *et al.* (Plenum, New York, 1991), p. 51.
[5] A. Mysyrowicz and C. Benoit à la Guillaume, Phys. Rev. Lett. **45**, 1970 (1980).
[6] V. B. Timofeev, V. D. Kulakovskii, and I. V. Kukushkin, Physica (Amsterdam) **117B&118B**, 327 (1983).
[7] N. Peyghambarian, L. L. Chase, and A. Mysyrowicz, Phys. Rev. B **27**, 2325 (1983).
[8] D. W. Snoke, J. P. Wolfe, and A. Mysyrowicz, Phys. Rev. Lett. **64**, 2543 (1990); E. Fortin, S. Fafard, and A. Mysyrowicz, Phys. Rev. Lett. **70**, 3951 (1993).
[9] L. V. Butov and V. D. Kulakovskii, Pisma Zh. Eksp. Teor. Fiz. **53**, 444 (1991) [JETP Lett. **53**, 466 (1991)]; L. V. Butov, V. D. Kulakovskii, G. E. W. Bauer, A. Forchel, and D. Grützmacher, Phys. Rev. B **46**, 12 765 (1992).
[10] Yu. E. Lozovik and V. I. Yudson, Zh. Eksp. Teor. Fiz. **71**, 738 (1976) [Sov. Phys. JETP **44**, 389 (1976)].
[11] T. Fukuzawa, E. E. Mendez, and J. M. Hong, Phys. Rev. Lett. **64**, 3066 (1990).
[12] J. A. Kash, M. Zachau, E. E. Mendez, J. M. Hong, and T. Fukuzawa, Phys. Rev. Lett. **66**, 2247 (1991).
[13] A. Zrenner, P. Leeb, J. Schäfer, G. Böhm, G. Weimann, J. M. Worlock, L. T. Florez, and J. P. Harbison, Surf. Sci. **263**, 496 (1992).
[14] I. V. Lerner, Yu. E. Lozovik, Zh. Eksp. Teor. Fiz. **80**, 1488 (1981) [Sov. Phys. JETP **53**, 763 (1981].
[15] G. E. W. Bauer, in *Optics of Excitons in Confined Systems*, IOP Conf. Ser. No. 123, (Institute of Physics, Giardini Naxos, Italy, 1991), p. 283.
[16] D. Yoshioka and A. H. MacDonald, J. Phys. Soc. Jpn. **59**, 4211 (1990).
[17] X. M. Chen and J. J. Quinn, Phys. Rev. Lett. **67**, 895 (1991).
[18] H. Hilmer, A. Forchel, R. Sauer, and C. W. Tu, Phys. Rev. B **42**, 3220 (1990).
[19] P. K. Basu and P. Rays, Phys. Rev. B **44**, 1844 (1991).
[20] V. N. Abakumov, V. I. Perel, and I. N. Yassievich, in *Nonradiative Recombination in Semiconductors*, edited by V. M. Agranovich and A. A. Maradudin (North-Holland, Amsterdam, 1991).
[21] B. L. Altshuler, D. E. Khmel'nitskii, A. I. Larkin, and P. A. Lee, Phys. Rev. B **22**, 5142 (1980).
[22] V. L. Nguen, B. Z. Spivak, and B. I. Shklovskii, Zh. Eksp. Teor. Fiz. **89**, 1770 (1985) [Sov. Phys. JETP **62**, 1021 (1985)].
[23] See, e.g., H. W. Jiang, C. E. Johnson, and K. L. Wang, Phys. Rev. B **46**, 12 830 (1992).
[24] B. L. Althshuler, P. A. Lee, and R. A. Webb, in *Mesoscopic Phenomena in Solids*, edited by V. M. Agranovich and A. A. Maradudin (North-Holland, Amsterdam, 1991).
[25] G. E. W. Bauer, Phys. Scr. **T45**, 154 (1992).
[26] E. I. Rashba and G. E. Gurgenishvilli, Fiz. Tverd. Tel. **4**, 1029 (1962) [Sov. Phys. Solid State **4**, 759 (1962)].
[27] J. Feldmann, G. Peter, E. O. Göbel, P. Dawson, K. Moore, C. Foxon, and R. J. Elliot, Phys. Rev. Lett. **59**, 2337 (1987).
[28] P. Dutta and P. M. Horn, Rev. Mod. Phys. **53**, 497 (1981).