Phase Transition of an Exciton System in GaAs Coupled Quantum Wells

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We have observed a sharp reduction of the photoluminescence linewidth from a two-dimensional exciton system (coupled GaAs/AlGaAs quantum wells), at a certain critical temperature (T_c) and under the influence of an electric field. We attribute this narrowing to a phase transition of the exciton system into an ordered state. The experiments suggest that a coherence length long enough to wash out the effect of the fluctuations, which cause the line broadening, is introduced below T_c.

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Bose-Einstein condensation (BEC) of excitons or excitonic molecules has been extensively discussed.[1] In bulk crystals, degenerate Bose-Einstein (BE) statistics for excitons[2] in Ge and biexcitons[3] in CuCl have been reported. Also, BE saturation[4] in Cu_2O has been observed. No similar observation in two-dimensional systems has been reported up to now.

One of the important factors that makes it difficult to show the direct evidence for BEC in any exciton system is the usually short excitonic lifetime. For example, Snoke and Wolfe have argued that the lifetime of excitons in Cu_2O is not sufficiently long compared with the thermalization time,[5] thus preventing the formation of a condensed state. The exciton lifetime of indirect-gap materials is, in general, longer than the thermalization time, but excitons condense into electron-hole plasma droplets before BEC.[1]

Although it is generally believed that in a strictly two-dimensional system, ideal BEC cannot occur, the Kosterlitz-Thouless transition can be expected. Indeed, this kind of transition has been studied both theoretically and experimentally in superconducting and ^{4}He films.[6]

For a two-dimensional system of spatially separated electron-hole pairs, Lozovik and Yudson[7] and Shevchenko[8] independently have suggested the possibility of a phase transition into an ordered state. Their proposal, based on an analogy with BCS theory, has recently been extended by Fukuzawa and co-workers to a coupled quantum-well system subject to an electric field, which allows the realization of a long-lived two-dimensional exciton gas.[9,10] In such a system, when a sufficiently strong electric field is applied perpendicular to the layers, electrons and holes become localized in different quantum wells, as shown in Fig. 1. The electrical polarization of excitons prevents the formation of molecules or droplets due to the effective alignment of electron-hole pairs in separate planes. In addition, since the overlap of the wave function can be varied easily by changing the applied field strength, the lifetime of excitons, which is inversely proportional to that overlap, can be controlled externally.

In this Letter we report photoluminescence (PL) experiments in coupled quantum wells (CQW) under an electric field that strongly suggest the existence of a phase transition of a two-dimensional exciton system to a collective state. The transition, manifested by a sharp reduction in the linewidth of the PL spectra, takes places below a critical temperature $T_c \simeq 10$ K, with an electric field of about 3 kV/cm.

The experiments were done in a heterostructure deposited by molecular-beam epitaxy on a n^+-type GaAs substrate (the growth proceeding without interruption between the various layers) and consisting of ten undoped double-well units separated by 200 Å of $Ga_{0.7}Al_{0.3}As$ from one another, with 5000 Å of heavily doped p^+-type GaAs on top. The units were formed by two 50-Å GaAs layers separated by 40 Å of $Ga_{0.7}Al_{0.3}As$ in between. Each of these independent units gives rise to a potential profile that, in the presence of an electric field $\mathcal{E}$ perpendicular to the layers, localizes electron and hole states in

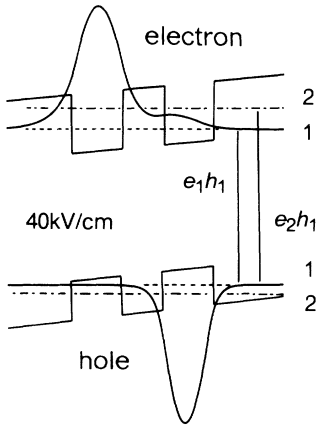

FIG. 1. Potential profile and electron and heavy-hole wave functions for the ground states of a coupled quantum well under an electric field. The width of the wells (GaAs) is 50 Å and that of the barrier (GaAlAs) is 40 Å.

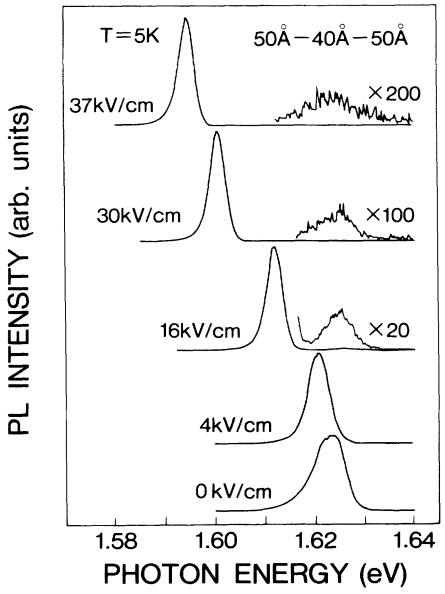

FIG. 2. Observed PL spectra for various electric fields at 5 K. At zero field the peak corresponds to the e_1h_1 transition of Fig. 1. As the field increased, this transition shifted toward lower energy and a new, weak peak, from the e_2h_1 and e_1h_2 transitions, was observable.

separate GaAs wells, as illustrated in Fig. 1. Electric fields, up to 40 kV/cm, were applied by means of a voltage between the n^+ and p^+ electrodes.

The sample was in contact with cold helium vapor inside a variable-temperature cryostat with optical access. The temperature, in the range 2–100 K, was measured with a calibrated Si diode and controlled by a heater to better than 0.1 K. Optical excitation, from 1.57 to 1.73 eV, was provided by a cw LD700 dye laser pumped by a Kr^+ laser. Excitation power densities were determined with the help of a TV camera to monitor the laser spot size at the sample surface.

In the absence of an electric field, at 5 K the PL consisted of a single peak at 1.6231 eV, with a full width at half maximum (FWHM) of 7.4 meV. This feature, shown at the bottom of Fig. 2, corresponds to the radiative recombination of electrons in the lowest, "symmetric," state of the conduction band with analog hole states of the valence band, indicated in Fig. 1 by e_1h_1. The other allowed interband transition at zero field (the "antisymmetric"-electron–"antisymmetric"-hole transition denoted by e_2h_2 in the nomenclature of Fig. 1) was also observed in photoluminescence excitation spectra, but will not be discussed in detail here.

Under an electric field the single PL peak shifted to lower energy and then split into two components, one of which shifted to lower energy with increasing field whereas the other initially moved to slightly higher energy and then remained unchanged (see Fig. 2). Simultaneously, the intensity of the former increased rapidly,

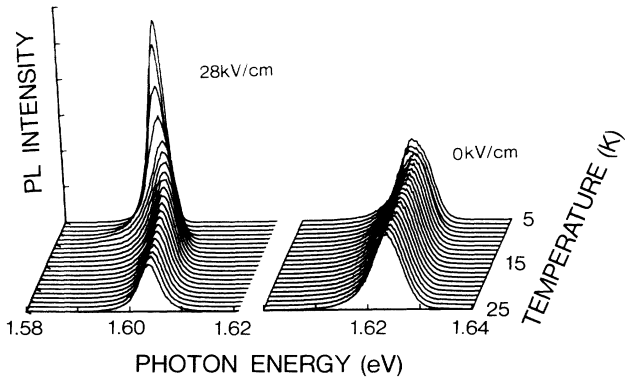

FIG. 3. Temperature dependence of the PL spectra at $\mathcal{E}=0$ and 28 kV/cm.

and then saturated, with increasing field, while the latter's strength decreased. The weak, high-energy peak corresponds to the e_2h_1 transition of Fig. 1, involving electron and hole states mostly localized in the right-hand-side well. On the other hand, the strong, low-energy peak represents a transition, e_1h_1, in which an electron and a hole are spatially separated in opposite wells. Because of the Coulomb interaction between them, this pair forms an exciton whose lifetime is determined by the small wave-function overlap. The field-induced energy shift of the e_1h_1 transition, which has been studied before,[11] is essentially proportional to $\mathcal{E}D$, where D is the separation between the centers of the quantum wells (90 Å). However, in our case there is an additional, extraordinary shift at moderate and high fields, *only* at low temperatures, which we will describe below.

A remarkable effect of the field is the drastic reduction of the PL linewidth at 5 K, which can be easily observed in Fig. 2 by comparing the spectra at 0 and 30 kV/cm, with FWHM of 7.4 and 3.4 meV, respectively. In patent contrast, the FWHM of the e_2h_1 PL line increased with increasing electric field. The most striking characteristic of the anomalous linewidth reduction is its strong temperature dependence (Fig. 3), which made the FWHM at 28 kV/cm drop abruptly from 5.6 meV at 9 K to 3.4 meV at 5 K. Such a change, summarized in Fig. 4, is most dramatic since it reverses the trend of a linewidth increase as the temperature decreases from 25 to 10 K.

The temperature-dependent transition is not limited to fields as high as 28 kV/cm. At lower fields a linewidth reduction is also observable, although the saturation of the linewidth occurs at temperatures below 5 K (the lower the field, the lower the temperature) and the transition becomes less sharp, as illustrated in Fig. 4. Finally, in the absence of field the linewidth increases monotonically with decreasing temperature.

Not only does temperature affect drastically the PL linewidth at a given field but also its peak energy, as seen in Fig. 3. At 28 kV/cm the peak shifted to lower ener-

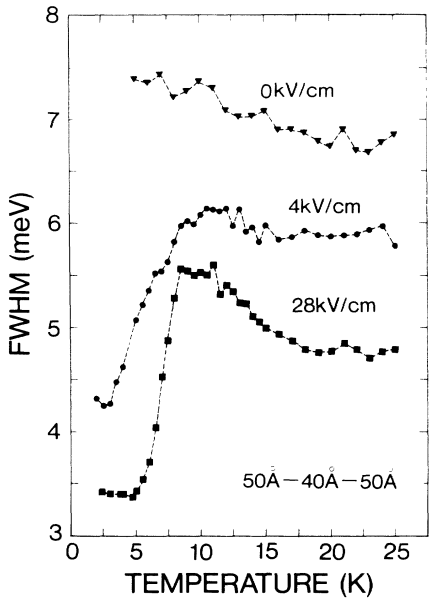

FIG. 4. Linewidth of the e_1h_1 transition as a function of the temperature at 0, 4, and 28 kV/cm. At 28 kV/cm, the width shows a sharp transition from 5.6 meV at 8.5 K to 3.4 meV at 5 K.

gies by more than 5 meV, from 25 to 5 K. At lower fields, the amount of shift was reduced, and for $\mathcal{E}=0$ it was only $\simeq 1.5$ meV. The zero-field shift can be easily understood in terms of two competing recombination mechanisms of free and bound excitons. At the lowest temperatures, excitons are bound to impurities or trapped at interface defects,[12] which a temperature increase sets free. Indeed, the observed shift of $\simeq 1.5$ meV is consistent with the binding energy of excitons to either impurities or defects and can be explained as a result of thermal activation. In contrast, at high fields the 5-meV shift is too large to be assigned to any thermally activated detrapping of excitons.

These PL effects depend on the excitation power density, but not on the excitation energy, at least in the range considered here—that is, between 1.65 and 1.72 eV. The results presented in Figs. 2 and 3 correspond to a density of $\simeq 0.58$ W/cm^2, for which the field-induced reduction of the linewidth was maximum. At either lower or higher densities, the line reduction was less pronounced. A detailed study of the temperature dependence of the linewidth for densities below the optimum value revealed a similar behavior to that of Fig. 4, although with more complicated PL line shapes above 10 K and with a saturation of the linewidth to values larger than 3.4 meV, and at temperatures below 5 K.

Photoluminescence excitation (PLE) spectra between 1.57 and 1.73 eV, for various fields, showed optical-absorption-like peaks corresponding to the e_1h_1 and e_2h_1 transitions. Not surprisingly, the intensity of the former was much weaker than that of the latter, even more so as the field increased, as a result of the decreasing wave-function overlap. The most relevant aspect of the spectra, however, was that, in contrast to the PL results, the linewidths of both PLE peaks remained unchanged, or even increased slightly, with increasing electric field.

We attribute the novel effects described above to a phase transition in the exciton system, from a normal regime to a collective regime under an electric field. To understand how the proposed transition can affect the PL linewidth it is first necessary to consider the origin of the spectral width in the absence of field. In wide quantum wells, the width of the PL line is mainly due to homogeneous broadening. On the other hand, in a system with narrow wells and barriers such as for these experiments, interface roughness caused by thickness fluctuations (which create lateral terraces in the layer planes) contributes significantly to an additional broadening.[13,14] Assuming a monatomic layer fluctuation (with an average thickness of 18-14-18 layers corresponding to the 50-40-50-Å structure), a computer simulation gave a line spread over $\simeq 20$ meV, with a spacing between individual lines of $\simeq 1$ meV. Experimentally, the observed spectrum has a FWHM of $\simeq 7$ meV with a tail that extends over $\simeq 20$ meV.

Tanaka and Sakaki[14] have concluded that whether the PL spectrum consists of a single, broad line or of several closely spaced lines depends on the size of the terraces relative to the excitonic diameter. Moreover, they have correlated the terrace size with growth conditions. If the epitaxial growth of the heterostructure is interrupted between layers for several seconds, the terraces are on the average much larger than the exciton radius ($\simeq 100$ Å) and individual narrow PL lines are observable. On the other hand, under uninterrupted growth the terrace size is comparable to that of the exciton and then the PL consists of a single broad line. Although we have not measured the terrace dimensions in our heterostructure, it is reasonable to assume that since the growth was not interrupted, terrace and exciton sizes are of the same order.

If the exciton system makes a transition to a state with long-range order, the excitonic energy levels will be determined by a potential averaged over an area much larger than the diameter of a single exciton. As a consequence, the PL broadening associated with interface roughness is suppressed and the linewidth is sharply reduced.

Such a phase transition in the CQW structure is made possible by the electric field that separates electron and hole wave functions, thus increasing drastically the exciton recombination time from a value estimated to be less than 0.5 ns. This short time may not be long enough for the excitons to thermalize before evolving into a condensed state,[5] since the number of collisions within that time may be less than a few hundred (using a velocity of 10^7 cm/s at 10 K). Our calculations show that at 28 kV/cm the lifetime increases by more than 2 orders of magnitude, which would make the transition much more

likely than in the absence of a field.

If indeed it is a long recombination time that makes the transition possible, a system with very strongly coupled quantum wells should be less favorable for the formation of a condensed state. In the other limit, of no coupling at all, spatially separated electrons and holes would not be able to form an exciton and therefore no transition should be observed. In addition to the heterostructure discussed here in detail, we have studied two other samples with similar parameters, except that the barrier between the quantum wells was either 50 or 20 Å. In the former case a field-induced narrowing was also present, although not as clear as in Fig. 2. On the other hand, when the barrier was 20 Å no line narrowing was observed, consistent with a short recombination time that would prevent the phase transition.

The true nature of the transition that our experimental results suggest is not known. An ideal Bose condensation is unlikely, given the quasi-two-dimensional character of our system. A BCS-like transition, as proposed by Fukuzawa and co-workers,[9,10] is possible; an estimation of the transition temperatures even gives a value of $\simeq 10$ K, consistent with our observation. However, such an agreement may be fortuitous since the experimental excitation density, estimated to be $\lesssim 10^{10}$ cm^{-2} from the pumping intensity, falls outside the range of validity of the theory.[15] If the transition is Kosterlitz-Thouless type, one would expect T_c to decrease as the exciton density decreases.[6] Unfortunately, from our present data it is difficult to give a precise value for the critical temperature. As noted above, the temperature at which the minimum linewidth is reached decreases as the excitation power density is decreased, but, on the other hand, the temperature for the onset of the linewidth reduction, $\simeq 10$ K, is not very sensitive to power-density variations.

More experiments are needed to further correlate PL linewidth with exciton condensation and to clarify the type of transition observed as well as the microscopic mechanisms that make it possible. Also, detailed calculations of collective effects in spatially separated electron-hole systems including recombination processes are necessary.

In summary, we report here the first observation of a phase transition in an exciton system consisting of spatially separated electron-hole pairs. We hope that future experimental and theoretical work will help clarify the precise nature of this phase transition.

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[1]For a review, see, e.g., P. Nozieres, Physica (Amsterdam) **117&118B**, 16 (1983).

[2]V. B. Timofeev, V. D. Kulakovskii, and I. V. Kukushkin, Physica (Amsterdam) **117&118B**, 327 (1983).

[3]N. Peyghambarian, L. L. Chase, and A. Mysyrowicz, Phys. Rev. B **27**, 2325 (1983).

[4]D. Snoke, J. P. Wolfe, and A. Mysyrowicz, Phys. Rev. Lett. **59**, 827 (1987).

[5]D. W. Snoke and J. P. Wolfe, Phys. Rev. B **39**, 4030 (1989).

[6]For a review, see, e.g., P. Minnhagen, Rev. Mod. Phys. **59**, 1001 (1987), and references therein.

[7]Y. E. Lozovik and V. I. Yudson, Pis'ma Zh. Eksp. Teor. Fiz. **22**, 556 (1975) [JETP Lett. **22**, 274 (1975)].

[8]S. I. Shevchenko, Fiz. Nizk. Temp. **2**, 505 (1976) [Sov. J. Low Temp. Phys. **2**, 251 (1976)].

[9]T. Fukuzawa, S. S. Kano, T. K. Gustafson, and T. Ogawa, Surf. Sci. **228**, 482 (1990).

[10]T. Fukuzawa, T. K. Gustafson, and E. Yamada, IEEE J. Quantum Electron. (to be published).

[11]M. N. Islam, R. L. Hillman, D. A. B. Miller, D. S. Chemla, A. C. Gossard, and J. H. English, Appl. Phys. Lett. **50**, 1098 (1987); H. Q. Le, J. J. Zayhowski, and W. D. Goodhue, Appl. Phys. Lett. **50**, 1510 (1987).

[12]G. Bastard, C. Delalande, M. H. Meynadier, P. M. Friklink, and M. Voos, Phys. Rev. B **29**, 7042 (1984); C. Delalande, M. H. Meynadier, and M. Voos, Phys. Rev. B **31**, 2497 (1985).

[13]H. Jung, A. Fischer, and K. Ploog, Appl. Phys. A **33**, 97 (1984).

[14]M. Tanaka and H. Sakaki, J. Cryst. Growth **81**, 153 (1987).

[15]Y. E. Lozovik and V. I. Yudson, Zh. Eksp. Teor. Fiz. **71**, 738 (1976) [Sov. Phys. JETP **44**, 389 (1976)].