Photoluminescence Ring Formation in Coupled Quantum Wells: Excitonic Versus Ambipolar Diffusion

M. Stern,* V. Garmider, E. Segre, M. Rappaport, V. Umansky, Y. Levinson, and I. Bar-Joseph
Department of Condensed Matter Physics, The Weizmann Institute of Science, Rehovot, Israel
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In this Letter, we study the diffusion properties of photoexcited carriers in coupled quantum wells around the Mott transition. We find that the diffusion of unbound electrons and holes is ambipolar and is characterized by a large diffusion coefficient, similar to that found in *p-i-n* junctions. Correlation effects in the excitonic phase are found to significantly suppress the carriers' diffusion. We show that this difference in diffusion properties gives rise to the appearance of a photoluminescence ring pattern around the excitation spot at the Mott transition.

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The lateral diffusion of indirect excitons in coupled quantum wells (CQW) has been a subject of intensive interest lately in the context of creating a cold and dense interacting exciton gas. The long lifetime [1] of these excitons allows them to diffuse for relatively long distances while cooling down to the lattice temperature [2]. Furthermore, the electrons and holes in this structure are typically separated by ~10 nm, giving rise to strong dipole moments, which are all aligned in the same direction. The interaction between the dipoles generates a strong force, which drives the excitons away from each other [3]. These properties have been used in a series of recent experiments, which attempted to reach the critical density and temperature for Bose-Einstein condensation (BEC) [4–7]. An interesting observation in these experiments is the appearance of two photoluminescence (PL) rings around the excitation spot. The external ring, which was originally considered as a manifestation of a degenerate exciton gas, was later shown to be due to recombination of excess free carriers in the quantum well [8,9]. The inner ring was attributed to enhanced luminescence of excitons as they cool down while diffusing away from the hot excitation spot [4]. A detailed hydrodynamic model was proposed to describe this diffusion, and concluded that a high degeneracy of the exciton gas has been achieved [10,11]. This was perceived as a step towards BEC, and an optical trap which is based on this effect was demonstrated [6,7].

In this Letter, we study the formation of the inner PL ring and show that the excited system is not a degenerate exciton gas but rather consists of unbound electrons and holes (*e-h*). We find that *e-h* diffusion is ambipolar and is characterized by a large diffusion coefficient, similar to that found in *p-i-n* junctions [12,13], while correlation effects in the excitonic phase significantly suppress the carriers' diffusion. Hence, there is a drastic change in the diffusion properties of the system at the Mott transition [14], and we show that it gives rise to the appearance of the ring pattern. The appearance of the ring can therefore be viewed as a manifestation of the exciton Mott transition in this system.

The sample is an *n-i-n* CQW structure grown by molecular beam epitaxy. The *i*-region consists of two GaAs quantum wells (QWs) of different widths (7/10 nm) separated by a 5 nm $Al_{0.28}GaAs$ barrier. Large area mesa (1.5 mm^2) and electrical contacts to the top and bottom layers were formed. The leakage current through the sample was substantially suppressed by etching most of the top *n* layer, leaving a thin resistive layer. The resulting dark current at 24 kV/cm was below 50 pA. The sample was mounted in a magneto-optic cryostat ($T = 1.6$ K), and was illuminated by a Ti:Sapphire laser, which was focused on a 15 μm diameter spot. A setup of motorized lenses collected the PL from different points of the sample, with a resolution of 3 μm. The laser energy was 1.56 eV, slightly above the wide QW gap but below the narrow well gap. Under these conditions, direct excitons are formed in the wide well only, and when an electric field of 24 kV/cm is applied, the electrons tunnel into the narrow well to form indirect excitons. The fact that the excitation energy is well below the $Al_{0.28}GaAs$ gap substantially reduces the photoleakage current (below 100 nA at 3 mW), and does not create excess free carriers in the QWs [8].

Figure 1 summarizes our experimental results at low excitation power. To determine the nature of the excited system, we measure the energy dispersion in a magnetic field B applied in Faraday configuration. In Fig. 1(a), we show the PL energy as a function of B at the center of the excitation spot, where the density is highest. One can clearly observe a quadratic behavior, which characterizes a bound exciton [15]. This quadratic behavior is observed at any measured location along the PL profile, supporting the conclusion that the diffusing carriers at this excitation power are bound indirect excitons.

In Fig. 1(b), we show the PL intensity profile of the indirect excitons. For comparison we also show the PL profile of the direct excitons, measured at zero electric field. It can be seen that the indirect excitons diffuse to a distance of ~10 μm, while the direct exciton diffusion length is at least an order of magnitude smaller. This large difference in diffusion length is a straightforward result of

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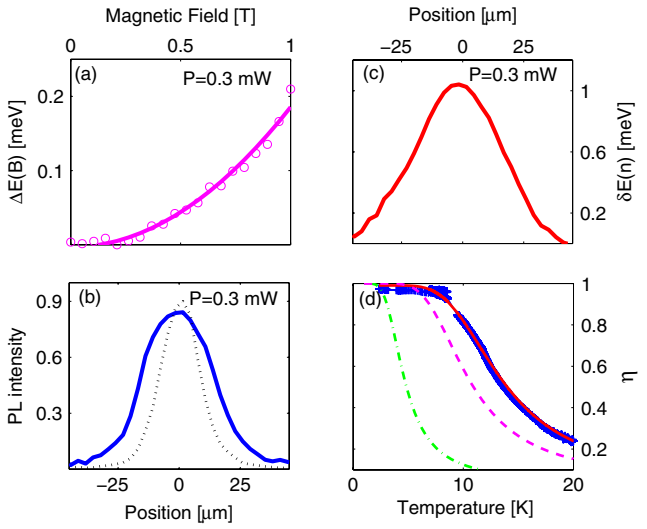

FIG. 1 (color online). (a) The PL energy shift as a function of B. The solid line is a fit to a quadratic diamagnetic behavior. (b) The PL intensity profile of the indirect exciton at $F = 24$ kV/cm (solid line). The dashed line is the direct exciton profile at $F = 0$, which effectively gives the spot size. (c) The PL energy shift as function of position. (d) The measured ratio $\eta = I_{IX}/(I_{IX} + I_X)$ as function of lattice temperature (crosses), and theoretical curves for $\delta E = 0$, 1 and 3 meV (solid, dash, dot-dashed lines, respectively).

their different lifetimes. Hence, to a good approximation the direct exciton intensity profile can serve as a measure of the spot size.

Figure 1(c) shows the PL energy shift, δE, as function of the distance from the center of the excitation point. This energy shift is a many-body effect due to the repulsive exciton-exciton interactions and depends on the local exciton density n. A theoretical description of this interaction was recently formulated [16,17], and it was shown that it can be expressed as

$$E_{\rm int} = \frac{ne^2 d}{\varepsilon} f(T) \tag{1}$$

where d the distance between the two well centers and ε the dielectric constant. The factor $f(T)$ is small at cryogenic temperatures, $f(2K) = 0.08$, and its origin is the strong depletion of the exciton gas around each exciton, which decreases the total energy of the system. An approximated expression for this correlation factor $f(T)$ can be obtained assuming a simple dipole-dipole interaction between excitons, $U_{dd}(r) = (2\pi\varepsilon)^{-1}(e^2/r - e^2/\sqrt{r^2 + d^2})$, and that the probability of finding an exciton at a distance r near a given exciton is $n \exp[-U_{dd}(r)/T] d^2\vec{r}$. One can then write the interaction as $E_{\rm int} = \int n U_{dd}(r) \times \exp[-U_{dd}(r)/T] d^2\vec{r}$, and for $T \ll e^2/(2\pi\varepsilon d)$, we obtain

$$f(T) \approx \Gamma(4/3) \sqrt[3]{\frac{\varepsilon d T}{8e^2}}. \tag{2}$$

This expression gives $f(T)$ values, which are very similar to the those obtained by Ref. [17].

The measured PL blueshift (relative to the very low power energy), $\delta E(n) \equiv E_{\rm int}$, at the spot center is $\sim$1 meV [Fig. 1(c)], corresponding to a total density $\sim 5 \times 10^{10}$ cm^{-2}. Clearly, the fact that the blueshift decreases with distance is a manifestation of the decrease of the steady state exciton density due to recombination and expansion of the exciton gas. We note, however, that the blueshift curve does not follow exactly the PL intensity curve. This is due to the slight heating of the lattice at the center of the excitation spot. We recall that the PL of an exciton gas is due to cold excitons, whose center of mass momentum is smaller than the photon momentum [18–20], and, hence, the PL from the hot region at the center is suppressed. To determine the local temperature at the spot center, we use the fact that the PL emission at this point consists of both direct and indirect exciton lines. The ratio $\eta = I_{IX}/(I_{IX} + I_X)$ between the PL intensities of these two lines is related to the temperature by

$$\eta = \frac{1}{1 + \gamma \exp\{[-\Delta E + \delta E(n)]/T\}} \tag{3}$$

where ΔE is the difference between the two exciton energies and γ is the ratio of their lifetimes. To calibrate the curve, we performed a measurement of η as a function of lattice temperature in a broad spot and under very low excitation power, where one could safely assume that the excitons are at lattice temperature and $\delta E(n) \approx 0$ [Fig. 1(d)]. It can be seen that the theoretical curve gives a good fit to the data with $\gamma = 300$ and $\Delta E = 6$ meV, in a good agreement with the expected values for these parameters. We find that at low powers [Fig. 1(c)] $I_X \ll I_{IX}$ at the spot center, implying $\eta \approx 1$. Using Eq. (3) and $\delta E(n) = 1$ meV, we conclude that $T < 5$ K. At high intensities, where $\delta E(n) = 2.5$ meV, we measure $\eta \approx 0.7$ and thus $T = 6$ K.

The spatial profile of the indirect excitons PL drastically changes at high excitation powers, and acquires a ring shape, which surrounds the excitation spot. The typical radius of this ring is $\sim$20 μm [Fig. 2(a)]. The formation of this ring was reported in a number of recent works and was interpreted in terms of exciton diffusion [4]. A model, which accounts for the exciton Bosonic nature, was formulated [11] and fitted to the experimental results. It was argued that the exciton diffusion coefficient increases dramatically near to the BEC transition temperature, and gives rise to the PL ring formation. We shall show below that the ring formation is not due to exciton diffusion but rather due to ambipolar diffusion of unbound electrons and holes, and is a vivid demonstration of the Mott transition.

Figure 2(b) describes the PL energy shift, $\delta E(n)$, as a function of distance from the spot center. It is seen that the energy profile is flat at the center, $\delta E \sim 2.5$ meV, and then decreases monotonically. If we assume that the diffusing particles are bound excitons and use Eq. (1) to determine their density, we are led to conclude that this blueshift corresponds to a very high exciton density, of

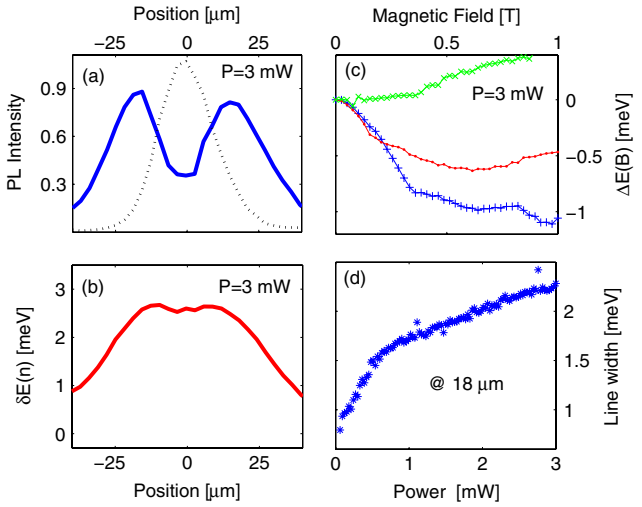

FIG. 2 (color online). (a) The PL intensity profile of the indirect (solid line) and direct (dashed line) excitons ($F = 24$ kV/cm and 0, respectively). (b) The PL energy shift as function of position. (c) The PL energy as a function of B at 3 points at distances of 18(+), 36(·), and 54(×)μm from the spot center. The zero for each of the curves is taken as the energy at zero magnetic field at each point. (d) The PL linewidth as a function of intensity at 18 μm from the spot center.

$\sim 1.25 \times 10^{11}$ cm^{-2}, well above the Mott transition in this system [14]. We note that even larger blueshifts were measured in Ref. [6], and were interpreted using an expression in which the $f(T)$ factor in Eq. (1) was omitted. The exciton density was therefore underestimated by an order of magnitude.

The key experiment to distinguish between e-h plasma and excitons is measuring the diamagnetic shift. We therefore spatially resolved the PL from various points across the diffusion profile and measured the PL peak energy as a function of B. Figure 2(c) shows the behavior at three points, at distances of 18, 36, and 54 μm from the spot center. It is seen that very far from the excitation spot, the behavior is diamagnetic, and a quadratic dependence is observed, indicating that the electrons and holes are bound and form excitons in this region. As we get closer to the center of the spot, this behavior is drastically changed: rather than an increase of the PL energy, we observe a redshift as a function of B. The origin of this redshift is the Lorentz force acting on charge particles, which suppresses their diffusion and reduces their steady state density far from the spot center. This reduction in density is manifested as a decrease of the local PL energy relative to the energy at $B = 0$, which overrides the diamagnetic blueshift. We studied the behavior of the PL energy versus B at various power densities, and found that this change from diamagnetic to ''paramagnetic'' behavior coincides with the appearance of the ring pattern. These results clearly show that at high powers, the diffusing particles are unbound electrons and holes. This conclusion is supported by the observation of a change of the behavior of PL width with intensity [Fig. 2(d)], which coincides with the appearance of the ring, and was shown to indicate a Mott transition [14].

While it is not surprising that excitons undergo a Mott transition at high power density, it is not obvious why this transition should be manifested as such a drastic change in the diffusion behavior. We show below that the diffusion of unbound e-h is much faster than that of bound pairs, by approximately an order of magnitude at cryogenic temperatures, and that the ring formation is due to giant ambipolar diffusion [12,13] of e-h plasma in CQW.

Let us begin with the indirect exciton diffusion, which is described by

$$\frac{\partial n}{\partial t} = -\vec{\nabla} \cdot \vec{j}_{IX} + S \tag{4}$$

$$= D_X \nabla^2 n + \vec{\nabla} \cdot (\mu_X n \vec{\nabla} E_{\text{int}}) + S \tag{5}$$

where D_X, and μ_X are the diffusion coefficient and mobility of the excitons, respectively, $\vec{j}_{IX}$ is the exciton current, and S is the rate of generation or annihilation of excitons. The first term in Eq. (5) describes a regular diffusion process due to a density gradient, while the second term is the one which is due to the interaction. To compare the two terms, we use the Einstein relation, $\mu_X = D_X/T$ (T is assumed to be higher than the BEC temperature), and express the exciton current as

$$\vec{j}_{IX} = -D_X(1 + E_{\text{int}}/T)\vec{\nabla} n. \tag{6}$$

It is seen that the interaction term is dominant when $E_{\text{int}} \gg T$, implying that interaction energies of a few tenths of meV are sufficient to overcome the regular diffusion at cryogenic temperatures.

Let us turn now to the diffusion of unbound electron and hole plasma. It is possible to write the two different diffusion-drift equations for electrons and holes as

$$\frac{\partial n_{e,h}}{\partial t} = D_{e,h} \nabla^2 n_{e,h} \pm e\vec{\nabla}.(n_{e,h} \mu_{e,h} \vec{\nabla} \varphi_{e,h}) + S \tag{7}$$

where $\varphi_{e,h}$ is the electrical potential in each well. An important property of this system is its local charge neutrality: the separation of different sign carriers costs electrostatic energy, and the system minimizes it by keeping equal local densities of electrons and holes, e.g., $n_e(x, y) = n_h(x, y)$. This neutrality will hold in CQW when d is much smaller than the diffusion length. Under this assumption, one can explicitly calculate the electrostatic term and get the ambipolar diffusion current

$$\vec{j}_A = -D_A(1 + E_{\text{elect}}/2T)\vec{\nabla} n \tag{8}$$

where $D_A = 2D_e D_h/(D_e + D_h)$ is the ambipolar diffusion coefficient and $E_{\text{elect}} = ne^2 d/\varepsilon$ is the electrostatic energy density of the e-h system. Comparing Eqs. (6) and (8), we find that the diffusion of unbound electron and holes is much faster than that of bound pairs, by a factor of $f(T)$, which amounts to approximately an order of magnitude at cryogenic temperatures [17].

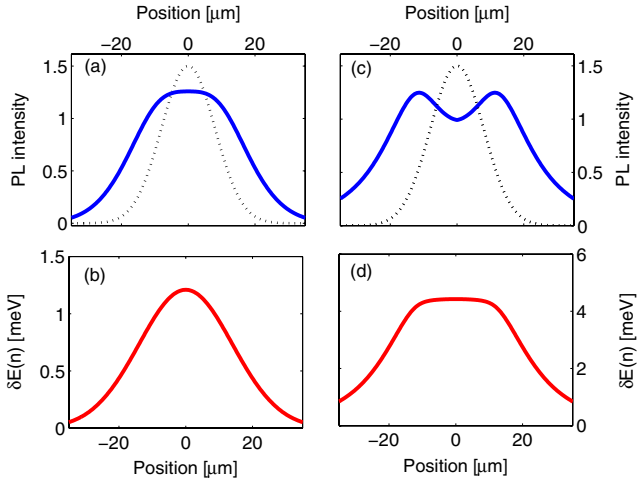

FIG. 3 (color online). Numerical solutions of the diffusion equations (5) and (7) for the PL intensity and energy shift as a function of position: (a), (b) the exciton case, and (c), (d) the e-h plasma case. A temperature gradient of 3 K which follows the spot profile (dashed line) is assumed in both cases. The excitation powers are 0.3 and 3 mW, respectively, the CQW absorption is 2%, $D_x = 3$ cm^2/s, and the indirect exciton lifetime is 30 ns.

To understand the formation of the ring, one should account for the temperature gradient from the central hot region at the spot center towards the sides. It is well known that the integrated PL intensity of e-h plasma goes like $\beta(T)n^2$, and it is easy to show that $\beta(T) \sim T^{-1}$ for a Maxwell-Boltzman distribution. Thus, the outer regions in the plateau of nearly constant density of Fig. 2(b) will emit more intensely than the regions at the spot center, giving rise to the ring structure. Figure 3 shows numerical solutions of Eqs. (4) and (7). It can be seen that we could reproduce the bell shape of the excitonic diffusion, the ring pattern of the PL intensity, and the plateau in the density profile in the case of ambipolar diffusion. We fitted the numerical results to the experimental data and found that the exciton diffusion coefficient is 3 cm^2/s, in agreement with the measurements of [2].

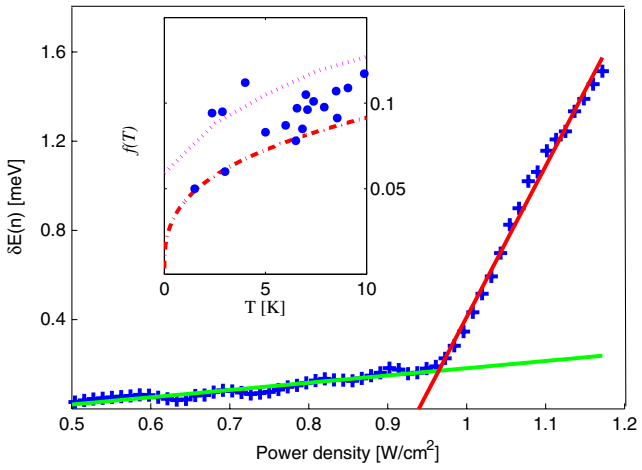

FIG. 4 (color online). The blueshift of the PL energy as a function of intensity in a broad spot (250 μm diameter) experiment around the Mott transition. The solid lines are linear fits in the exciton and e-h regimes. The ratio of their slopes yields $f(T = 1.5\ \mathrm{K}) = 0.05$. (inset) The measured values of the correlation factor f as function of temperature. The lines are the theoretical calculation of Ref. [17] (dashed line) and Eq. (2) (dot-dashed line).

A central point in our analysis is the dependence of the interaction energy on density, Eq. (1). In the concluding part of this Letter, we wish to experimentally determine the value of $f(T)$. In Fig. 4, we show the blueshift of the PL energy as a function of intensity in a broad spot (250 μm diameter) experiment. The PL was collected from a small area of a few tens of microns in diameter, ensuring that the exciton density in the area from which we collect the PL is constant. The change in slope at the Mott transition is clearly visible and is due to the different density dependence of the PL energy, $\delta E(n)$, in the two sides of the transition. Using the expressions for E_{int} and E_{elect}, we can use the measurement to determine the correlation factor, $f(T)$. We find that $f(1.5\ \mathrm{K}) = 0.05$ and that its temperature dependence is very close to the prediction of Ref. [17].

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*mstern@weizmann.ac.il

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