# Mott Transition of Excitons in Coupled Quantum Wells

M. Stern,* V. Garmider, V. Umansky, and I. Bar-Joseph

*Department of Condensed Matter Physics, The Weizmann Institute of Science, Rehovot, Israel*

(Received 2 November 2007; revised manuscript received 19 April 2008; published 23 June 2008)

In this work we study the phase diagram of indirect excitons in coupled quantum wells and show that the system undergoes a phase transition to an unbound electron-hole plasma. This transition is manifested as an abrupt change in the photoluminescence linewidth and peak energy at some critical power density and temperature. By measuring the exciton diamagnetism, we show that the transition is associated with an abrupt increase in the exciton radius. We find that the transition is stimulated by the presence of direct excitons in one of the wells and show that they serve as a catalyst of the transition.

DOI: 10.1103/PhysRevLett.100.256402 PACS numbers: 71.35.Lk, 73.21.Fg, 78.67.De

The exciton Mott transition [1] is seemingly a straightforward prediction for the behavior of an interacting electron-hole system. At low temperature, most of the electrons and holes should be bound in excitons. As the density (or temperature) increases, more and more excitons ionize, releasing free carriers. The free carriers screen the Coulomb interaction that binds the remaining excitons, facilitating their ionization. Further increase of the density (temperature) leads to avalanche ionization. This thermodynamic transition, which is also called the ''ionization catastrophe'' [2], has attracted a lot of attention and controversy in the past several decades. Indeed, conflicting theoretical predictions have been recently published, with some predicting an abrupt phase transition at a critical temperature or density [3], while others argue that there should be a gradual transition [4]. The experimental data seem to reflect this controversy: While the experiment of Kappei *et al.* in GaAs quantum wells shows a gradual transition [5], Amo *et al.* provide evidence for an abrupt transition in bulk GaAs [6].

Indirect excitons ($IX$s) in coupled quantum wells (CQWs) offer a unique opportunity to revisit this longstanding problem. These excitons are formed by electrons and holes that reside in different quantum wells (QWs), separated by a thin tunnel barrier. Their close proximity allows them to interact and form a bound state. This unique structure gives rise to far-reaching consequences. In particular, the $IX$ lifetime can be made very long, as long as microseconds, limited only by the exponentially small overlap of the electron-hole ($e$-$h$) wave functions [7]. This lifetime is much longer than the relaxation time with the thermal bath of the lattice [8–10] and the exciton formation time [11–14] and thus allows us to establish a thermodynamic equilibrium, alleviating a central problem in direct exciton experiments.

In this work we study the phase diagram of optically generated $IX$s in a CQW system and show that it undergoes a phase transition to an unbound electron-hole plasma. This transition is manifested as an abrupt change in the photoluminescence (PL) linewidth and peak energy at some critical power density and temperature. By measuring the exciton diamagnetism, we show that the transition is associated with an abrupt increase in the exciton radius, from 20 nm below the critical density to >50 nm above it. We find that the transition is stimulated by the presence of direct excitons in one of the wells and show that they serve as a catalyst of the transition. Indeed, we find that in the absence of these catalysts the phase transition is not observed.

The sample that we investigated is an $n^+$-$i$-$n^+$ structure grown by molecular beam epitaxy on a semi-insulating GaAs substrate. The $i$ region consists of two GaAs quantum wells of different width, 7 and 10 nm, separated by a 5 nm $Al_{0.28}GaAs$ barrier and surrounded by two AlGaAs spacer layers. An electric field of 24 kV/cm was applied in a direction perpendicular to the QWs, such that the electron level in the wide well is higher than that of the narrow well. Changing the strength of this field modifies the $e$-$h$ overlap integral and therefore the carrier's recombination time. To perform the PL measurements, we prepared a large area sample (1.5 mm$^2$). The mesa was illuminated by a laser spot which covered approximately a quarter of the area, and the PL was collected from a small spot of a few tens of microns in diameter. This ensures that the exciton density in the area from which we collect the PL is uniform.

The samples were excited by using a tunable Ti:sapphire laser. By selecting the excitation energy $E_L$ to be above or below the narrow well gap $E_{NW}$, we could select two different initial conditions. At $E_L < E_{NW}$, electrons and holes are excited in the wide well only. The electrons quickly tunnel into the narrow well, and at steady state one gets nearly complete charge separation: The electrons reside in the narrow well, while the holes are in the wide well. The only excitons that are observed under this condition are indirect ones. This is changed when $E_L > E_{NW}$, when electrons and holes are created at both wells. Here also the electrons tunnel into the narrow well. However, the tunneling time of the holes is much longer, and we get a population of holes trapped at the narrow well. As a consequence, we get a mixture of two types of excitons:

 

indirect and direct ones, the latter being formed between electrons and holes in the narrow well. We shall see that this difference has far-reaching implications on the nature of the transition. We note that in both cases the excitation energy is below the $Al_{0.28}GaAs$ gap; thus, the leakage current is significantly reduced relative to the recently reported He-Ne experiments [15–17]: A typical value is 250 $\mu A/cm^2$ at 1 $W/cm^2$ (at an electric field of 24 kV/cm). We verified that the accumulation of extra charges in the QWs, originating from shallow impurities in the spacer, is negligible [18].

Let us begin with the case of $E_L < E_{NW}$ at $T = 1.5$ K, where only indirect excitons can be formed. The binding energy of the indirect excitons is $E_B = 2.5$ meV, 20 times larger than $k_B T$ at that temperature. Hence, the number of ionized excitons is expected to be negligibly small, suppressed by a factor of $\exp(\frac{-E_B}{k_B T})$ relative to the bound ones. Indeed, the PL measurements show that the carriers are bound in excitons even at very high densities. In Fig. 1(a), we show the PL energy shift as a function of power density. We observe a relatively small blueshift of 0.6 meV as the power density is increased up to 4 $W/cm^2$. The origin of this blueshift is the interaction and correlations between the excitons, which have a permanent dipole along the growth direction and, thus, exercise a repulsive force on each other. By using the recent calculations of Zimmermann and Schindler [19], one can estimate the exciton density that corresponds to the observed blueshift to be $6 \times 10^{10}$ $cm^{-2}$. This density is comparable with the so-called Mott density $1/a_B$, where $a_B$ is the exciton Bohr radius, yet no transition is observed. The presence of a high exciton density is also manifested in the PL spectrum: The linewidth increases by 50%, from 1 to 1.5 meV over this density range [Fig. 1(b)], while the integrated intensity grows precisely linearly with power.

To get a deeper insight into the nature of the excited system, we measured the behavior of the PL energy as a

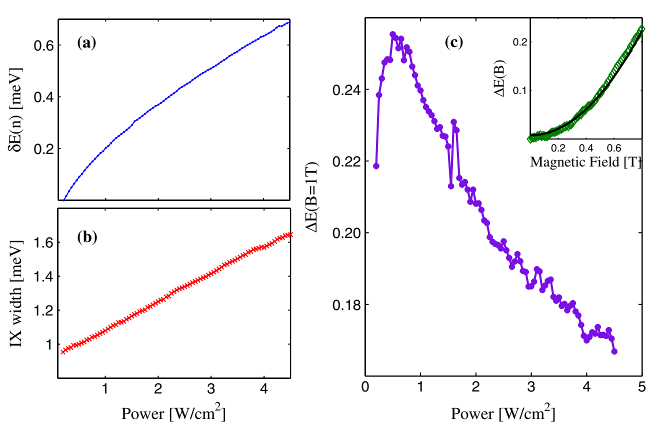


FIG. 1 (color online). The PL properties for excitation in the wide well only $E_L < E_{NW}$ $T = 1.5$ K. (a) The PL energy shift, (b) the linewidth, and (c) the diamagnetic coefficient of the *IX* peak as a function of power density. Inset in (c): Quadratic behavior of the peak energy as a function of the magnetic field.

function of magnetic field $B$, applied in a Faraday configuration. It is well known that the exciton ground state energy should increase quadratically with magnetic field, a phenomenon known as the exciton diamagnetic shift [20]. The shift is given by $\Delta E(B) = \alpha B^2 \langle r^2 \rangle$, where $\langle r^2 \rangle$ is the expectation value of the exciton radius squared, $\alpha = \frac{e^2}{8\mu c^2}$, and $\mu$ is the reduced mass of the exciton in the plane parallel to the QWs. This is in contrast to an uncorrelated $e$-$h$ plasma, which exhibits a linear dependence of the energy on $B$, given by $\Delta E(B) = \hbar\omega_c/2$, where $\omega_c$ is the cyclotron frequency. This measurement allows us to distinguish between excitons and plasma and to determine the expectation value of the exciton radius $\sqrt{\langle r^2 \rangle}$. The inset in Fig. 1(c) shows a typical measurement of PL energy dependence on $B$. A clear parabolic dispersion is seen throughout this density range, supporting the conclusion that the carriers are bound and form excitons. We find that the diamagnetism *decreases* with increasing exciton density, indicating that the exciton radius becomes smaller at high density. This is in clear contrast with the naive Mott scenario, where the excitons are expected to be less bound as the density increases. Such behavior has been predicted by Koch *et al.* and was attributed to segregation of the carriers into separate domains of bound and unbound electron-hole pairs [4].

Let us turn now to the second case of $E_L > E_{NW}$, where we get a mixture of direct and indirect excitons. We find that the dependence of PL on power density is radically different, and an abrupt change is found at 0.75 $W/cm^2$. Figure 2(a) shows the change of the linewidth near the transition: An abrupt change from a width of 1 meV below the transition to 1.2 meV above it is seen. It is interesting to note that the transition is accompanied by a modification of the line shape from being predominantly Lorentzian at low power densities to Gaussian at high densities. The transition is extremely abrupt and cannot be resolved within our power resolution (0.1 $mW/cm^2$). Figure 2(b) shows a close-up view of the behavior of the PL energy versus power near the transition point: It can be clearly seen that the slope of the curve abruptly changes from shallow to steep. To understand the origin of this behavior, let us consider the density dependence of the PL energy in two limits, an uncorrelated $e$-$h$ plasma and an exciton gas. In the $e$-$h$ plasma regime, the separation of charges in the two wells creates an electric field which screens out the external electric field and shifts the recombination energy to higher values. The blueshift of the PL energy can be straightforwardly calculated as $\delta E(n) = \frac{4\pi e^2 d}{\varepsilon} n$, where $n$ is the density of the uncorrelated $e$-$h$ pairs, $d$ is the separation between the centers of the two wells, and $\varepsilon$ is the dielectric constant. On the other hand, at the excitonic regime the energy shift is primarily due to the repulsive interaction and correlations between the excitons, as discussed earlier. In a simplified picture, each exciton creates a depletion region around itself, and as a consequence the

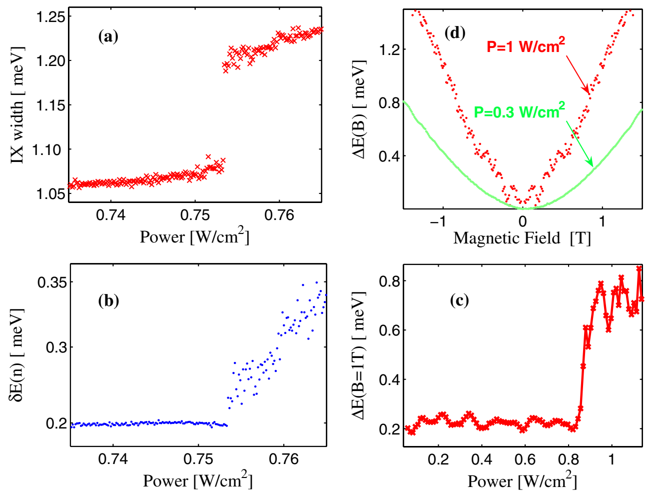


FIG. 2 (color online). The PL properties for excitation in both wells $E_L > E_{NW}$ $T = 1.5$ K. (a) The linewidth, (b) the PL energy shift, and (c) the diamagnetic coefficient as a function of the power density. (d) The diamagnetic shift at two power levels, below and above the transition. The magnetic field dependence measurements were conducted in a different cryostat with fiber illumination.

blueshift is temperature-dependent and much weaker than in the uncorrelated case. The blueshift-based estimate of the exciton density at the transition [19] is $2 \times 10^{10}$ cm$^{-2}$ at 1.8 K. To confirm this estimate, we performed PL measurements at magnetic fields and determined the magnetic field at which the lowest Landau level (LL) is full. At this magnetic field, the PL from the second LL is diminished. This measurement can be conducted only at high carrier densities, well above the transition. We find that the carrier density depends linearly on power, and we could therefore extrapolate the linear dependence to extract the density at the transition. This estimate agrees very well with that based on the blueshift.

Here again we obtain a deeper insight by measuring the diamagnetic behavior. Figure 2(d) shows the PL energy as a function of magnetic field for two power densities, below and above the transition. One can clearly see how the quadratic behavior at low power densities turns into almost linear at high power. The in-plane radius of the indirect excitons at low power can be readily extracted from this measurement and found to be ~20 nm. Applying this analysis to the high power data yields a lower bound of 50 nm for the exciton radius. We verified the validity of this approach by comparing it to the diamagnetic shift of the direct exciton, which is found to yield the expected radius of ~10 nm. This clearly indicates that the transition is accompanied by a substantial change of exciton size, from being bound to unbound. Figure 2(c) shows the diamagnetic coefficient as a function of power density. Two regimes of diamagnetism are clearly observed, with a sharp boundary between a phase of bound excitons and a phase of unbound *e-h* pairs. It is interesting to note that we do not observe a gradual increase of the exciton radius below the transition as implied by the Mott transition scenario.

To establish the fact that this is indeed a thermodynamic phase transition, we measured the temperature dependence of the transition. Figure 3(a) shows the evolution of the PL linewidth and blueshift with temperature at $P = 0.95$ W/cm$^2$. We find that at low temperatures ($T = 1.6$ K) the linewidth is broad and reaches 1.8 meV, characteristic of an unbound electron-hole plasma. As the temperature is increased, the width decreases rapidly and levels at 1.3 meV above 3 K. Upon further increase of the temperature, we observe an opposite transition above 6 K—the PL linewidth turns from excitonic to *e-h* plasma. The same reentrance behavior is observed when examining the blueshift of the PL energy. The phase diagram in the power-temperature ($P$-$T$) plane is depicted in Fig. 3(b) [the dashed line describes the measurement of Fig. 3(a)]. One can see that the excitonic phase indeed exists only in a limited area in this parameter space.

Our results clearly show that in the case of a mixed $X$ and $IX$ phase an abrupt thermodynamic phase transition takes place, between bound and unbound *e-h*. A natural question to ask is what the underlying mechanism is that distinguishes this system and gives rise to the observed behavior. An insight into the origin of this mechanism is obtained by carefully examining the changes in the direct exciton luminescence. We find that at the phase transition threshold a new line appears 2.1 meV below the narrow well $X$ line [Fig. 4(a)], and its intensity increases linearly at higher powers. By examining its behavior in electric and magnetic fields, we could unambiguously identify it as the narrow well trion $T$, a bound state of two electrons and a hole [21,22].

To understand the implications of this observation, we should recall that the carriers are arranged such that all electrons reside in the narrow well while the holes are divided between the two wells. This situation is schematically depicted in Fig. 4(c). Clearly, these carriers can lower their energy by forming bound states, and two configurations can be identified: The first is forming a mixture of

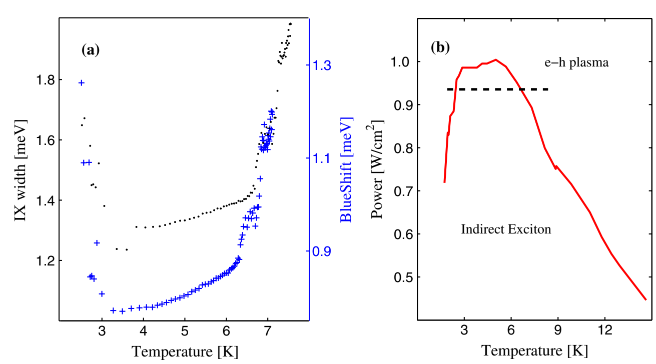


FIG. 3 (color online). (a) The linewidth (points) and blueshift (crosses) as a function of temperature for $P = 0.95$ W/cm$^2$. (b) Power-temperature phase diagram showing the $IX$ and *e-h* regions. The dashed line corresponds to the parameters of (a).

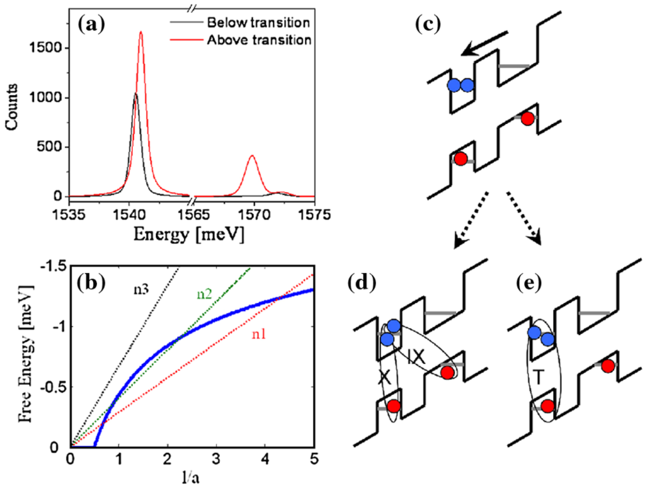


FIG. 4 (color online). (a) PL spectra below and above the transition showing the large increase in the $T$ line above the transition. (b) The free energy of screened excitons (solid line) and an electron-hole plasma (dashed lines) for different total pair densities as a function of the screening length $l$. (c)–(e) Energy band diagram of the coupled quantum wells under a perpendicular applied electric field showing the charge separation and the possible bound states.

direct and indirect excitons $X + IX$ [Fig. 4(d)], and the second is a trion and a free hole $T + h$ [Fig. 4(e)]. To compare the relative energies of these two configurations, it is useful to consider the case where direct excitons are already present and calculate the energy gain by forming each configuration. In the first case, we gain 2.5 meV by binding the electrons and holes in the two wells into indirect excitons, while in the second case only 2.1 meV is gained by forming a trion and a free hole. Clearly, the second configuration is higher in energy; hence, the $X + IX$ is the favorable configuration at low density. The energy difference between the two configurations is, however, only 0.4 meV, which is 2–$3k_BT$ in the temperature range of the experiment. Therefore, a substantial portion of the $IX$ ionizes to $T$ through the process $IX + X \rightarrow T + h$. Since their lifetime is very short, the trions recombine quickly and leave behind free electrons. Unlike the pure $IX$ case, where the ionization level is a binding energy above, we have here an efficient channel of ionizing the $IX$, which is only a few $k_BT$ higher than the ground state. This allows us to set the necessary conditions for the ionization catastrophe to occur and reproduce the conditions for the Mott transition to be observed.

In the concluding part of this Letter, we wish to return to the $P$-$T$ phase diagram and suggest a qualitative explanation for its unique shape. Let us consider two states of the system: a bound exciton gas and a free $e$-$h$ plasma. If we assume that the screening of the excitonic interaction is due to free $e$-$h$, and it can be treated within the Thomas Fermi approximation, we may write the free energy per particle of the exciton system as $f_X = U_B(l/a_B) - s_xT$, where $U_B$ is the screened binding potential, $l$ is the screening length, and $s_x$ is the entropy. In the Thomas Fermi approximation $l = \frac{2\varepsilon k_BT}{e^2 n}$, where $n$ is the free $e$-$h$ density, and $-R_y \leq U_B \leq 0$ for $1/2 \leq l/a_B \leq \infty$. The solid line in Fig. 4(b) shows some generic dependence of $U_B$ on $l$ or, equivalently, on $T/n$. In the other limit of the $e$-$h$ plasma, the free energy can be written simply as $f_{e\text{-}h} = -s_{e\text{-}h}T$. The ground state of the system can therefore be determined by comparing $f_X$ and $f_{e\text{-}h}$, and the transition line is determined by solving $f_X - f_{e\text{-}h} = 0$. A graphical solution is shown in Fig. 4(b) for several $e$-$h$ densities (for the sake of clarity, $s_XT$ was substracted from $f_X$ and was added to $f_{e\text{-}h}$). It is seen that this equation has two solutions for a wide range of values and no solution beyond a certain density, as indeed observed experimentally. While this model provides a qualitative understanding of the transition, it is very simplified: It considers the exciton screening within a simple approximation and neglects changes in the exciton distribution [4] which should modify the system's entropy. It also neglects interaction effects and band gap renormalization in the $e$-$h$ plasma. These require a more detailed analysis, which is beyond the scope of the current Letter.

The authors thank Y. Levinson, B. Laikhtman, and D. Snoke for fruitful discussions. This research was supported by the Israeli Science Foundation, the Minerva foundation, and the Scientific Cooperation Program of the French Embassy in Tel Aviv.

*mstern@weizmann.ac.il


[1] N. F. Mott, *Metal Insulator Transitions* (Taylor & Francis, London, 1990).
[2] T. M. Rice *et al.*, Solid State Phys. **32**, 1 (1977).
[3] S. Ben Tabou de Leon and B. Laikhtman, Phys. Rev. B **67**, 235315 (2003).
[4] S. W. Koch *et al.*, Phys. Status Solidi B **238**, 404 (2003).
[5] L. Kappei *et al.*, Phys. Rev. Lett. **94**, 147403 (2005).
[6] A. Amo *et al.*, J. Appl. Phys. **101**, 081717 (2007).
[7] J. E. Golub *et al.*, Phys. Rev. B **41**, 8564 (1990).
[8] T. Fukuzawa, E. E. Mendez, and J. M. Hong, Phys. Rev. Lett. **64**, 3066 (1990).
[9] J. A. Kash *et al.*, Phys. Rev. Lett. **66**, 2247 (1991).
[10] V. B. Timofeev *et al.*, Phys. Rev. B **61**, 8420 (2000).
[11] T. C. Damen *et al.*, Phys. Rev. B **42**, 7434 (1990).
[12] D. Robart *et al.*, Solid State Commun. **95**, 287 (1995).
[13] C. Piermarocchi *et al.*, Phys. Rev. B **55**, 1333 (1997).
[14] S. Siggelkow *et al.*, Phys. Rev. B **69**, 073104 (2004).
[15] L. V. Butov *et al.*, Nature (London) **417**, 47 (2002).
[16] D. Snoke *et al.*, Nature (London) **418**, 754 (2002).
[17] C. W. Lai *et al.*, Science **303**, 503 (2004).
[18] S. I. Gubarev *et al.*, JETP Lett. **72**, 324 (2000).
[19] R. Zimmermann and C. Schindler, Solid State Commun. **144**, 395 (2007).
[20] M. Bugajski, W. Kuzko, and K. Reginski, Solid State Commun. **60**, 669 (1986).
[21] G. Finkelstein, H. Shtrikman, and I. Bar-Joseph, Phys. Rev. Lett. **74**, 976 (1995).
[22] I. Bar-Joseph, Semicond. Sci. Technol. **20**, R29 (2005).