Excitonic Mott transition in double quantum wells

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(Dated: August 28, 2003)

We consider an electron-hole system in double quantum wells theoretically. We demonstrate that there is a temperature interval over which an abrupt jump in the value of the ionization degree occurs with an increase of the carrier density or temperature. The opposite effect — the collapse of the ionized electron-hole plasma into an insulating exciton system — should occur at lower densities. In addition, we predict that under certain conditions there will be a sharp decrease of the ionization degree with increasing temperature — the anomalous Mott transition. We discuss how these effects could be observed experimentally.

PACS numbers: 71.35.-y, 71.35.Ee, 73.21.Fg

It was suggested by Mott [1] that an increase of the temperature or density of an insulating system of excitons may lead to avalanche ionization as a result of screening and k-space filling. Despite significant efforts [2, 3, 4, 5, 6], there is still no firm theoretical understanding of this effect, and the existence of the transition is disputed.

In bulk indirect-bandgap semiconductors, experimental observation of the Mott transition is prevented by the formation of electron-hole liquid (EHL) droplets [7]. The absence of low-temperature phase transitions in direct semiconductors and single quantum wells is believed to be caused by fast radiative recombination, which does not allow enough time for the photo-generated electron-hole plasma (EHP) to cool down.

In recent years there has been increased attention towards double quantum wells (DQWs) — heterostructures containing two quantum wells (QWs) situated close enough so that the Coulomb correlations between particles in different QWs are significant. Electrons and holes from adjacent QWs form spatially indirect excitons (IXs) with very long lifetimes, due to the small electron-hole (e-h) wavefunction overlap. The experimentally confirmed long life-time and stability of the DQW indirect exciton [8, 9, 10, 11, 12] suggests that the formation of a medium-density cold exciton gas is possible. Under certain conditions this gas may undergo a Mott transition.

Another property beneficial for observation of the Mott transition in a DQW is that the spatially separated EHP possesses substantial electrostatic energy (Hartree term in the self-energy). This leads to the fact that for large enough separation of quantum wells there is no minimum in the dependence of the ground-state energy on the density, and thus the formation of e-h droplets is not possible [5, 13].

The purpose of this Letter is to investigate the possibility of a Mott transition in DQW systems and to encourage further experimental research.

The main theoretical difficulty in investigation of the Mott transition is that it is inherently a medium-density effect, for which low-density and high-density approximations do not work. The correct approach would require careful handling of the Coulomb interaction depending on the thermodynamic state of the system, which implies a self-consistent procedure.

Before studying the Mott transition, which is essentially a jump in the ionization degree of the EHP, one should clarify the concept of ionization degree and exciton density at medium and high plasma densities. In the dilute case [14], excitons can be considered as well-defined Bose particles, and the problem of finding the ionization degree can be solved simply by counting electrons and holes in bound states pairwise. This approach, however, runs into trouble when the density of carriers increases and the exciton wavefunctions start to overlap. Furthermore, this simple chemical picture is not applicable if the scattering states are occupied, since counting particles in scattering states pairwise leads to divergences.

Therefore, it appears more practical to formulate the theory in terms of primary quasi-particles, i.e. in terms of electrons and holes. Instead of considering excitons in the system we investigate the single-particle density at given temperature $T = 1/\beta$ and quasi-Fermi energies $\xi_e$ and $\xi_h$. To this end we use many-body theory for a system of interacting quasi-particles within the ladder approximation. Starting with the self-energy in the quasi-particle ladder approximation, which is based on the electron-electron (e-e) and e-h statically screened pair-interaction series, after a lengthy derivation [15] similar to that of Zimmermann and Stolz for the 3D case [16], we obtain an expression for the electron density in the following form:

$$n_e = n_e^0(\xi_e) + n_{ee}(\xi_e) + n_{eh}(\xi_e + \xi_h),$$

where $n_e^0 = m_e/(\beta\pi\hbar^2)\ln(1 + \exp(\beta\xi_e))$ is the density of free quasi-particles, and $n_{ee}$ ($n_{eh}$) originates from the e-e (e-h)
(e-h) interaction:

\[
n^{ab} = \frac{2}{\beta \pi \hbar^2} \sum_{m=-\infty}^{+\infty} \left[ (1 - \delta_{ab}) \sum_n M_{eh} L_{eh} (\epsilon_{mn}) + \frac{1}{M_{ab} \lambda_m^2} \int_0^\infty dk L_{ab} \left( \frac{h^2 k^2}{2m_{ab}} \right) 2 \sin^2 \frac{\delta_{ab}}{2} \left( \frac{d\delta_{ab}}{dk} \right) \right],
\]

(2)

Here, \( m_e (m_h) \) is the electron (hole) effective mass, \( M_{ab} = m_a + m_b, m_{ab} = m_a m_b / (m_a + m_b), \lambda_m = 1 - \delta_{ab} (-1)^m / 2 \), and \( L_{ab}(\epsilon) = -\ln [1 - \exp (\beta (\xi_a + \xi_b - \epsilon))] \). The exciton levels \( \epsilon_{mn} \) and generalized phase shifts \( \delta_{m}^{ab} \) are obtained as the poles and the complex arguments of the \( m \)th Fourier components of the T-matrix. The T-matrix equation

\[
T^{ab}(\mathbf{q}, \mathbf{q}', \Omega_{ab}) = -V_s^{ab}(\mathbf{q}' - \mathbf{q}) - \sum_{\mathbf{q}''} V_s^{ab}(\mathbf{q}' - \mathbf{q}) \frac{1 - f_a(q'') - f_b(q' - q'')}{\epsilon_a(q'') + \epsilon_b(q' - q'') - \Omega_{ab}} T^{ab}(\mathbf{q}'', \mathbf{q}', \Omega_{ab})
\]

(3)
is solved using the factorized-potential approximation \[12, 13\]. Here, \( \epsilon_{(h)} \) are the electron (hole) quasi-particle energies and \( f_{(h)}(\epsilon) = [\exp (\beta (\epsilon - \xi_{(h)}))] + 1 \) are Fermi distribution functions. The major advantage of the mass action law for quasi-two-dimensional (2D) systems, in the form of Eqs. 1-3, over previously used approaches \[14\] is that it is not confined to low densities and accounts for k-space filling. It is essential to include the effects of k-space filling into a theory of the Mott transition since in two dimensions any weak symmetric potential has a bound state.

The next effect which should be carefully dealt with is screening. Using the Green’s function technique, we have derived the following general expressions for the linearily-screened Coulomb interaction in quasi-2D systems \[17\]:

\[
V_s^{ab} = \eta_a \eta_b v(q) \left[ F^{ab}(q) - v(q)\pi^{ab}(q, \Omega) S(q) / D(q, \Omega) \right].
\]

(4)

Here, \( v = 2\pi e^2 / q \) is the bare Coulomb potential, \( F^{ab} \) are the form factors, \( \pi^{ab} \) are the polarization functions, \( \eta_{(h)} = 1 (-1), \epsilon(h) = \hbar (\epsilon), S = F^{ee} F^{hh} - (F^{eh})^2, D = 1 - v \Pi_1 + v^2 \Pi_2 S, \) where \( \Pi_1 = \pi^{ee} F^{ee} + \pi^{hh} F^{hh} - 2\pi^{eh} F^{eh}, \) and \( \Pi_2 = \pi^{hh} \pi^{ee} - (\pi^{eh})^2 \).

Retaining only the ladder diagrams in the expressions of polarization functions \[18\], and using an approximation similar to the plasmon-pole approximation for the RPA polarization function \( \pi^{RPA} \), the following expressions are obtained: \( \pi^{ee} = \pi^{RPA} + \pi^{e\gamma}, \pi^{hh} = \pi^{e\gamma}, \pi^{eh} = \pi^{e\gamma} \), where

\[
\pi^{e\gamma}(q, \Omega) = -\frac{\hbar^2 q^2}{M_{ab} \left( \frac{2\pi e^2 \hbar^2}{m_{ab} q_s} q^2 + \frac{\kappa^2}{4 M_{ab} q^4} \right)} + \hbar^2 q^2 / \Omega^2,
\]

(5)

with the screening parameter

\[
q_s^{ab} = (1 + \delta_{ab}) \left( \frac{2\pi e^2}{\kappa} \right) \frac{d n^{ab}}{d(\xi_a + \xi_b)}.
\]

(6)

Inspection of Eq. 4 shows \[17\] that in spatially separated systems screening by IXs is of the same order as screening by the free-carrier plasma, and this is due to the well-defined dipole momentum of the exciton. This questions the validity of applying previously developed theories of quasi-2D screening, which ignore screening by IXs, to DQW systems.

In the quasi-particle approximation, the chemical potential \( \mu \) is given as the sum of the Fermi energy and the average quasi-particle shift \( \Delta_{(h)} \). Eqs. 1-3 show that the density is a function of the Fermi energies only (the relation between \( \xi_e \) and \( \xi_h \) is established by the charge neutrality). However, it is the quasi-particle shifts (sometimes called the exchange-correlation part of the chemical potential) that can lead to negative values of \( d\mu / dn \), which in this case will mean the formation of droplets. As was already mentioned, for large enough separations the Hartree term of the self-energy prevents a first-order e-h (or exciton) liquid phase transition; in this case \( \mu \) is a monotonic function of the quasi-Fermi energy and the shifts can be completely ignored \[19\]. This justifies the omission of the exciton-exciton interaction in the derivation of Eq. 1, since it will mainly contribute to the positive quasi-particle shifts, which are due to dipole-dipole repulsion.

The e-h correlation term of the total density, \( n_{eh} \), in Eqs. 1 and 2 depends on the sum of \( \xi_e \) and \( \xi_h \), and thus it is unchanged with a variation of the local field. This represents insulating behavior, whereas the first two terms in Eq. 1 depend on \( \xi_e \) alone and show metallic behavior. Therefore, it is reasonable to introduce the ionization degree
of the EHP as \(\alpha = 1 - n^{e\!h}/n_e = 1 - n^{e\!h}/n_h\), yielding a mass action law which is not restricted to the low-density regime.

We use a self-consistent procedure to calculate the parameters of the spatially-separated EHP. Screening is considered statically, i.e. \(\Omega = 0\) is assumed in Eq. (4). Fig. 1 shows the dependence of the free quasi-particle \((n^0)\), e-h \((n^{e\!h})\) and total \((n^{tot} = n^0 + n^{ee} + n^{e\!h})\) density in the DQW (the width of the QWs is 0.25 \(a_B^*\) and the distance between them is 0.2 \(a_B^*\)) on the electron quasi-Fermi energy. Here, \(a_B^*\) and \(Ry^*\) are the Bohr radius and Rydberg of the bulk (three-dimensional) exciton. The corresponding dependence of the ionization degree is plotted in Fig. 2. One can see that for high temperature (0.5 \(Ry^*\)) the total density increases monotonically, whereas in the case of low temperature (0.2 \(Ry^*\)) there is a region of values of the Fermi energy in which a growing free quasi-particle density is unable to compensate for the decrease of the correlated e-h component, \(n^{e\!h}\). In this region the total density curve acquires a negative slope, and a local maximum \((n^{M1})\) and minimum \((n^{M2})\) of the total density as a function of \(\xi_e\) appear. The ionization degree dependence takes a 'hysteresis' form (Fig. 2).

If the density of the e-h pairs in the system \(n^{tot}\) lies between \(n^{M1}\) and \(n^{M2}\), there are three corresponding states of the system characterized by three different Fermi energies, \(\xi_1\), \(\xi_2\) and \(\xi_3\) (see Figs. 1b and 2). The thermodynamically stable state is \(\xi_1\) since it has the smallest free energy (and ionization degree, see Fig. 2). The intermediate state \(\xi_2\) is unstable due to a negative compressibility, \(d\mu/dn = (d\xi_e/dn)(d\mu/d\xi_e) < 0\). The state \(\xi_3\) with the highest \(\alpha\) is quasi-stable, with a local minimum of free energy at the given density and chemical potential.

As the density gradually increases from zero, at \(n^{tot} = n^{M1}\) there will be an abrupt transition to the state with higher \(\xi\), shown in Figs. 1b) and 2 by a thick dash-dot arrow. Physically this situation means the following. Increasing the density of the carriers in the DQW system, one reaches a critical value \(n^{M1}\), at which a slight increase of the carrier concentration results in a large jump in the ionization degree. Effectively, this means a transition from a system consisting mainly of excitons to an almost completely ionized state. Clearly, this is what the Mott transition (avalanche ionization) is meant to be. There will be no phase separation as this first-order transition takes place between phases with different chemical potentials, i.e. equilibrium phase co-existence is impossible.

However, there is another transition mechanism. If the initial state of the system is a high-density ionized EHP and the density is slowly decreased, then the abrupt transition into an insulating state (ionized EHP collapse) may happen not at the Mott density \(n^{M1}\), but at a lower density \(n^{M2}\) (the thick dotted arrow in Figs. 1 and 2). The reason for the difference between these two densities can be explained qualitatively as follows. When the system is in the insulating state, the screening due to excitons is comparatively weak, and accumulation of a substantial exciton density is required in order to trigger the avalanche ionization. After the avalanche ionization occurs, the screening

**FIG. 1:** Dependence of the different components of the carrier density in a DQW on the electron quasi-Fermi energy.
FIG. 2: Ionization degree for an EHP in a DQW at two different temperatures. Arrows show possible phase transitions.

FIG. 3: The phase diagram for a DQW. The QW width is 0.25 $a_B^*$, and the distance between QWs is 0.2 $a_B^*$.

in the system is mainly due to free carriers, i.e. it is much stronger, and one has to step back in density much further in order for the system to collapse into excitons.

In Fig. 3 the phase diagram of a DQW structure is shown. Both the critical temperature $T_c$ and density $n_c$ are experimentally accessible ($T_c$ is about 15 K and $n_c$ about $2 \times 10^{10}$ cm$^{-2}$ for typical GaAs/AlGaAs structures). These parameters depend on the material and width of the individual QWs, as well as the interwell separation. As the temperature is decreased, the avalanche ionization density increases drastically (we introduce the characteristic minimum temperature $T_{min}$ as the temperature at which $n^{M1} = 1.0 \ a_B^{-2}$). The reason for this is that for lower temperatures the free-carrier density is reduced, and the weak screening by excitons and k-space filling requires higher densities to start the avalanche ionization process.

The phase diagram area below the transition curves corresponds to the situation when the system can reside in one of two states: a stable excitonic phase or metastable ionized EHP. The excitation conditions, and the direction and speed of the variation of the pumping intensity and temperature influence the final state. It is interesting to consider the transition from a metastable ionized EHP to excitons (or back) by crossing the low-density transition curve with
FIG. 4: The critical density (a) and temperature (b) as a function of e-h separation. In (b) the binding energy of the IX and the characteristic minimum temperature are also shown.

temperature variation. In this case, a sharp increase of excitonic luminescence with a slow increase in temperature or sharp disappearance of the excitonic line with a decrease of temperature can take place. This direction of the transition is opposite to the normal Mott transition, and so we call it the anomalous Mott transition.

In Fig. 4 the dependence of the critical parameters of the transition on the carrier separation is examined (the distance $d$ is measured between the centers of the QWs, and the points for a single-QW case, $d = 0$, are shown for the sake of comparison.) The critical density $n_c$ is decreasing with increasing e-h separation, which can be attributed to the increase of the IX radius. The critical temperature $T_c$ follows closely the decreasing trend of the IX binding energy $\epsilon_0$ (for large enough separation, $T_c \approx 0.25\epsilon_0$). The temperature $T_{min}$ is much less sensitive to the e-h separation. As a result, the temperature interval in which avalanche ionization can occur shrinks with increasing distance between QWs.

Encouraged by theoretical findings \[13, 20\], most experimental groups have been trying to obtain a quantum condensate of IXs \[8, 9, 10, 11, 12\]. In most cases this means concentrating the research effort on localized states and extremely low (below 1-3 K) temperatures. As we showed for delocalized excitons, the observation of the normal Mott transition at such temperatures is difficult due to large critical density. At temperatures below $T_{min}$, the steady enhancement of IX luminescence with an increase of pumping could be interpreted as a precursor to Bose condensation in some experiments.

Some experimental data \[12\] show that localized states become filled at densities less than $3 \times 10^9$ cm$^{-2}$, in which case delocalized excitons start to dominate photoluminescence spectra. The same source reports the observation of the insulator-metal transition at densities less than $10^{11}$ cm$^{-2}$. Unfortunately, no information on the sharpness of the transition and the temperature dependence of critical density has been provided.

Timofeev et al. \[8\] observed a sharp disappearance of the excitonic peak with a decrease of the temperature at temperatures $2K < T < 9K$ and low to moderate densities. In our opinion, this may be interpreted as the anomalous Mott transition. The measured transition curve is qualitatively similar to the anomalous Mott transition curve in Fig. 4.

We hope to encourage more experimental research which would lead to irrefutable observation of such a fundamental effect as excitonic Mott transition. Our theoretical results suggest the following method. The temperature should be set as low as possible and the pumping slowly increased to a high value, but so that the temperature kept constant. Then, as the system is placed in the stable insulating state below the normal Mott transition curve in Fig. 2, one should slowly increase the temperature. The Mott transition will manifest itself as the abrupt elimination of the excitonic peak from the luminescence spectra or an increase of the longitudinal photo-conductance. Once the ionized state is achieved, one can try to observe the anomalous enhancement of exciton luminescence with a decrease of the density (photo-excitation).
We wish to thank I. Galbraith for stimulating discussions and D.G.W. Parfitt for valuable advice.

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