Exciton Condensate in Semiconductor Quantum Well Structures

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We propose that the exciton condensate may form in a well-controlled way in appropriately arranged semiconductor quantum well structures. The mean-field theory of Keldysh and Kopaev, exact in both the high density and low density limits, is solved numerically to illustrate our proposal. The electron-hole pairing gap and the excitation spectrum of the exciton condensate are obtained. The energy scales of the condensate are substantial at higher densities. We discuss how such densities could be achieved experimentally by generating an effective pressure.

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The issue of Bose-Einstein condensation of excitons has been extensively studied, following the early suggestion by Keldysh and Kopaev [1–3]. In the intervening years, it continues to be a subject of considerable experimental and theoretical interest [4–10]. Until recently, much of the experimental work has been carried out on indirect semiconductors [1]. In view of the great advances made in our abilities to design and manufacture high quality artificial semiconductor quantum well (QW) structures, it appears opportune to investigate in some detail the possible exciton condensate states in direct gap semiconductors.

The crux of the matter lies in obtaining an exciton fluid at sufficiently high densities and low temperatures to realize a condensed phase. Recent experiments on Cu_2O [10] and on GaAs quantum wells in high magnetic fields [9] appear quite promising. However, existing work on QWs has usually been done in the low density regime, where it has been argued that disorder dominates the photoluminescence spectrum [8]. Clearly it would be very desirable to be able to produce exciton fluids at *controlled*, and possibly higher, densities. This has two advantages: the energy scale of the condensate would be larger and the variation of key properties with density could be systematically examined. Other fundamental questions related to the dynamics of condensation, possible lasing action, superradiance, and coupling to a coherent photon field could be studied experimentally under controlled conditions. Our proposal is to tailor the QW parameters in double well electron-hole systems so as to generate an effective pressure on part of the exciton fluid and thereby achieve the physical conditions necessary for a controlled formation of exciton condensates.

To this end, we set up and numerically solve the mean-field theory (MFT) proposed earlier by Keldysh and Kopaev [2,5,6], exact in both the high and low density limits, to obtain the total energy as a function of arbitrary densities for a variety of interlayer separation of the double layer QWs. The same MFT is also used to study the excitation properties of the proposed exciton droplet, which are crucial to its stability against small perturbations such as finite temperature, interface disorder, and complications arising from the band structure of the underlying semiconductors. At $T = 0$ the exciton fluid for the idealized system is an insulating Bose-Einstein condensate at all densities, i.e., there exists a gap to all charged excitations. Hence it has been called an excitonic insulator.

We begin by examining the ideal two-dimensional (2D) electron-hole system, characterized by the following Hamiltonian:

$$H_{eh} = \sum_i \frac{\vec{P}_{i,e}^2}{2m_e} + \sum_i \frac{\vec{P}_{i,h}^2}{2m_h} + \sum_{i<j} \frac{e^2}{\varepsilon|\vec{r}_{i,e} - \vec{r}_{j,e}|}$$
$$+ \sum_{i<j} \frac{e^2}{\varepsilon|\vec{r}_{i,h} - \vec{r}_{j,h}|} - \sum_{i,j} \frac{e^2}{\varepsilon\sqrt{|\vec{r}_{i,e} - \vec{r}_{j,h}|^2 + d^2}}, \tag{1}$$

where ε is the background dielectric constant. We ignore the electron-hole exchange in H_{eh} [1]. We assume that the electron and hole layers are infinitesimally thin, and that they are separated by a distance d [11,12]. Within the mean-field theory described later and for s-wave pairing between an electron and a hole, spin degrees of freedom play no role other than changing r_s by a factor of $\sqrt{2}$ at a given pair density n and will be ignored henceforth. In this work, the unit mass m is taken to be twice the reduced mass of the electron (m_e) and the hole (m_h), or $2/m = 1/m_e + 1/m_h$. The unit length is $a_{a.u.} = \varepsilon e^2/m\hbar^2$, and the energy unit is 1 Hartree $= e^2/\varepsilon a_{a.u.}$. r_s is defined as $\pi r_s^2 a_{a.u.}^2 = 1/n$.

Two obvious limits of this system are $r_s \to 0$ and $r_s \to \infty$. In the former high density limit, the pairing between the electrons and holes is weak and can be described in a BCS-like language where only properties near the Fermi surfaces are affected [2,5,6,13]. In the latter low density limit, excitons are formed and they interact weakly [3]. It has been realized that even in the latter limit, a BCS-like description is also appropriate [2,5,6]. In fact, it is exact in the limit of $r_s = \infty$, where we have single isolated excitons.

Such a formulation is also valid for 2D, which we will use in this work. The variational ansatz for the ground state wave function, taking into account the pairing effects, is in the BCS form [2,5,6,13]

$$|\Psi^0\rangle = \prod_{\vec{k}}[u_{\vec{k}} + v_{\vec{k}}a^\dagger_{e,\vec{k}}a^\dagger_{h,\vec{k}}]|\text{vac}\rangle, \tag{2}$$

with $|u_{\vec{k}}|^2 + |v_{\vec{k}}|^2 = 1$. $u_{\vec{k}}$ and $v_{\vec{k}}$ are the variational parameters to be determined for the present system, and $|\text{vac}\rangle$ is the vacuum state in which the valence band is completely filled, $a^\dagger_{e,\vec{k}}$ creates an electron in the conduction band, and $a^\dagger_{h,\vec{k}}$ creates a hole in the valence band. As in BCS, this wave function does not preserve the particle number. The solutions are obtained most easily by going to the second quantized form and minimizing the free energy at a given chemical potential μ, $f = \langle h_{eh}\rangle - \mu\langle n\rangle$, where lower cases are used to denote that the quantities are per unit volume. We find

$$\langle h_{eh}\rangle = \sum_{\vec{k}} \epsilon_{\vec{k}}v_{\vec{k}}^2 - \sum_{\vec{k}_1,\vec{k}_2}\left(\frac{1}{2}V^{ee}_{\vec{k}_1-\vec{k}_2}v_{\vec{k}_1}^2 v_{\vec{k}_2}^2\right.$$
$$+ \frac{1}{2}V^{hh}_{\vec{k}_1-\vec{k}_2}v_{\vec{k}_1}^2 v_{\vec{k}_2}^2$$
$$\left. + V^{eh}_{\vec{k}_1-\vec{k}_2}u_{\vec{k}_1}v_{\vec{k}_1}u_{\vec{k}_2}v_{\vec{k}_2}\right), \tag{3}$$

and $\langle n\rangle = \sum_{\vec{k}}v_{\vec{k}}^2$. In Eq. (3), $V^{ee}_q = V^{hh}_q = 2\pi/q$ and $V^{eh}_q = 2\pi e^{-qd}/q$. $\epsilon_{\vec{k}} = k^2/m$, the kinetic energy of an electron-hole pair, each with momentum $\vec{k}$. Notice that for $d \neq 0$ and in cases where the electrons and holes are not separately neutralized by the respective dopants (e.g., when they are created by optical pumping), the density-dependent Hartree term in the total energy needs to be included. At a fixed density, it increases energy/pair by $2d/r_s^2$ and μ by $4d/r_s^2$, without affecting the gap function Δ_k. Setting $\partial f/\partial v_{\vec{k}} = 0$ and considering only s-wave pairing in which case all quantities are functions of the magnitude of $\vec{k}$, we find in close analogy to the BCS algebra,

$$\xi_k = \epsilon_k - \mu - \sum_{\vec{k}'}V^{ee}_{\vec{k}-\vec{k}'}(1 - \xi_{k'}/E_{k'}), \tag{4}$$

$$\Delta_k = \sum_{\vec{k}'}V^{eh}_{\vec{k}-\vec{k}'}\Delta_{k'}/E_{k'}, \tag{5}$$

$$E_k^2 = \xi_k^2 + \Delta_k^2. \tag{6}$$

E_k is identified as the pair excitation spectrum [14].

Equations (4)–(6) are coupled equations that must be solved self-consistently. Given a chemical potential μ, we wish to find the corresponding density parameter $r_s = \sqrt{1/\pi n}$ and the energy per pair $E^{\text{pair}} = \langle h_{eh}\rangle/\langle n\rangle$. Letting $k = \tan(\beta)$, we set up a Gaussian-quadrature grid for β and convert the above equations into a matrix form which can be solved iteratively [15]. Only the angular averages of the potentials V^{ee} and V^{eh} enter, and the logarithmic singularities in the averaged quantities are treated separately [15]. The total energy per unit volume

is

$$\langle h_{eh}\rangle = \frac{1}{2}\sum_{\vec{k}}\left[(\epsilon_k + \mu + \xi_k)\frac{1 - \xi_k/E_k}{2} - \Delta_k\frac{\Delta_k/E_k}{2}\right]. \tag{7}$$

Shown in Fig. 1 are E^{pair} (solid lines) and μ (dashed lines) as a function of r_s from the present MFT calculation, for $d = 0$ and $d = 1.0$. For large r_s, E^{pair} and μ approach the correct respective single exciton binding energy, largest at $d = 0$. For small r_s, E^{pair} approaches the Hartree-Fock result, $2/r_s^2 - 16/3\pi r_s + 2d/r_s^2$, where the terms are the kinetic energy, the exchange energy, and the Hartree energy, respectively. Indeed, the present MFT approach describes both limits correctly in a natural way, and is expected to provide a reasonable interpolation for intermediate densities.

It is interesting to examine the strength of the pairing effects and the excitation spectrum of the exciton fluid. At a given layer separation d, the maximum in the gap function Δ_k (or the minimum in the excitation energy E_k) first increases with r_s, reaches a peak value, and then decreases with r_s. Its k dependence exhibits some interesting features as the chemical potential μ crosses the bottom of the single-particle band [15]. As an illustration, we show for $r_s = 2.66$ and $r_s = 5.90$ [16] at $d = 1.0$ the gap function Δ_k and the pair-excitation spectrum E_k as a function of k in Fig. 2.

First of all, E_k remains significant, in fact, ~0.4 Hartree, or ~7 MeV in GaAs QWs, at both of these densities. Second, we note that the gap function Δ_k exhibits a variation with k, instead of being sizable only very near k_F as in the BCS case. This is a direct result of the extended k range of the attractive interaction V^{eh}. Last, a comparison of the gap function at the two densities shows some qualitative differences between them. For the high density $r_s = 2.66$ case, the minimum
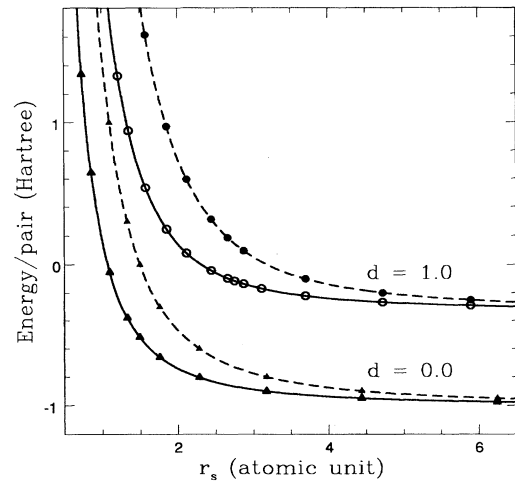

FIG. 1. Energy per electron-hole pair (E^{pair}, solid lines) and the chemical potential (μ, dashed lines) of a double-layer electron-hole system as a function of r_s at separations $d = 0$ and $d = 1.0$.
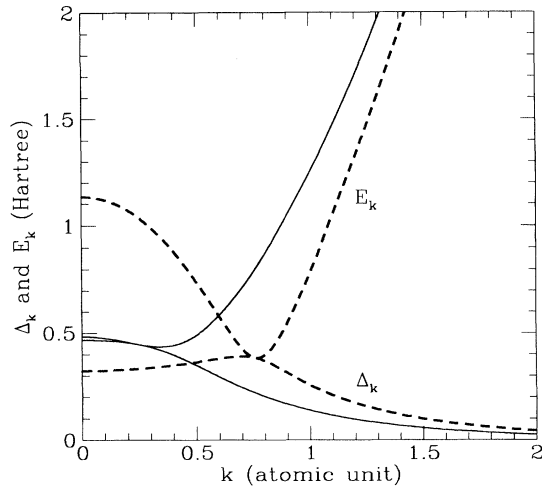
FIG. 2. Gap function Δ_k and the pair-excitation energy E_k of the exciton droplet as a function of k for $r_s = 2.66$ (dashed lines) and for $r_s = 5.90$ (solid lines) at $d = 1.0$.

pair-excitation energy in E_k is located very close in k to where the maximum in Δ_k occurs, and both are close to, but not identical to k_F. But for the low density $r_s = 5.90$ case, the maximum of Δ_k is now at k very close to zero, and the minimum of E_k is at k smaller than k_F [15]. Although we have emphasized the similarities between the present electron-hole problem and the BCS superconductivity, there are essential differences even at the level of MFT [15].

Screening effects will reduce both the gap function Δ_k and the minimum excitation energy in E_k. But we expect the calculated excitation energies to be qualitatively correct, since the very presence of the gap will make screening less effective on the energy scale of the gap itself. The basic gap structure of the excitation spectrum should certainly survive temperatures of a few K.

In the mean-field theory, Fig. 1 shows that the interaction between the excitons is repulsive so that only the gas phase exists in equilibrium in the uniform, double QW electron-hole system [17]. Such systems have been utilized in some recent photoluminescence experiments, and suggestive results for Bose-Einstein condensation have been reported, but only in the presence of a strong magnetic field at low densities [8,9]. We now show that an altered structure may induce an effective pressure on the exciton fluid and thereby stabilize higher densities.

We consider QW structures with a spatially varying well width, given by $w = w(\vec{r})$. Within the lowest subband approximation, the effects of $w(\vec{r})$ can be described by an effective in-plane potential $V(\vec{r}) = V(w(\vec{r}))$. If $dV/dr \ll V/d$, at equilibrium the exciton distribution throughout the QW $n(\vec{r})$ will be given by

$$\mu(n(\vec{r})) + V(\vec{r}) = \mu, \qquad (8)$$

where μ is the chemical potential of the excitons in the QW and $\mu(n)$ is the chemical potential of a uniform excitonic insulator at density n. Since the kinetic energy in the

z direction depends rapidly on w, changing w is a very effective way of controlling the local chemical potential $\mu(n(\vec{r}))$, thereby controlling the local density $n(\vec{r})$.

This is illustrated schematically in Fig. 3, where the well width on one side of the double QW is greater over a circle of radius R (the "cavity") than that of the rest. Only one side needs to be adjusted, since charge neutrality demands that the electron and hole densities be equal. A bias electric voltage is applied to spatially separate the electrons and holes after optical pumping. Several nanoseconds after pumping, during which the electrons and holes in the individual single layers have all recombined, we are left with a double-layer electron-hole system.

Notice that, for large r_s, the chemical potential $\mu(n)$ remains very close to the single exciton binding energy $|E_{ex}|$ for a wide range of density (see Fig. 1). Therefore, if the pumping intensity is not too great, the chemical potential of the excitons will remain at μ slightly larger (smaller in magnitude) than $-|E_{ex}|$. At equilibrium, the density inside the cavity n_{cav} will then be well approximated by $\mu(n_{cav}) = -|E_{ex}| + |\delta V|$. For GaAs/AlGaAs QWs, $m_h = 0.4$, $m_e = 0.067$, and $\varepsilon = 13$, we find the present atomic units have length $a^*_{a.u.} = 59.8$ Å and energy 1 Hartree $= 18.5$ meV. As an example, we consider the case where the hole layer width is adjusted. In order to change the local chemical potential inside the cavity from $-|E_{ex}|$ to $\mu(r_s = 3.69)$ at $d = 1$ (a value between the two r_s's considered earlier), the well widths can be arranged to be $w(\vec{r}) = 60$ Å for $r > R$ and $w(\vec{r}) = 73$ Å for $r < R$. We note that, quite generally, any QW structure similar to that of Fig. 3 will serve to
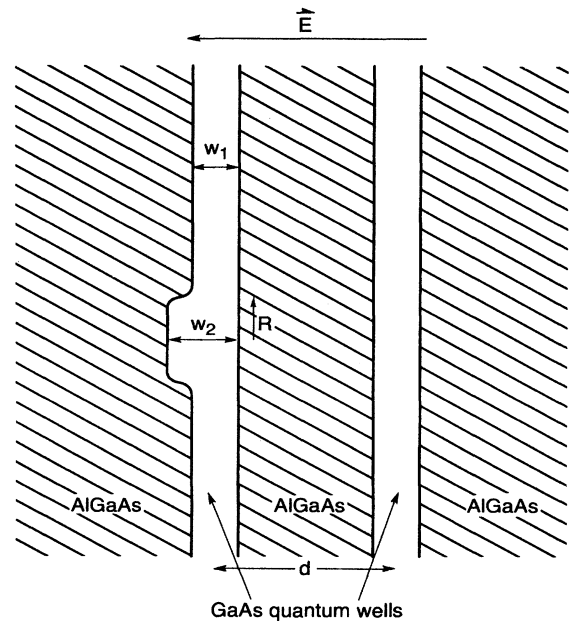

GaAs quantum wells

FIG. 3. Schematic diagram of the GaAs/AlGaAs quantum well structure expected to contain the exciton droplet at chemical equilibrium.

form an exciton droplet at a density largely controlled by the difference in the respective $w(\vec{r})$.

In order to obtain a Bose condensate excitonic insulator, the temperature must be sufficiently low and the lifetime of the excitons long enough. Currently attainable experimental conditions [8,9] satisfy the requirements: The lifetime can be extended to ~ 100 ns, due to the smaller electron-hole wave function overlap in the double layers; consequently, thermal and chemical equilibrium down to 1 K temperatures can be reached. Even if some of the excitons may be trapped at the impurity centers invariably present, a droplet of high density fluid should still form, and be insensitive to the trapping. One significant advantage of our proposal is that the properties of the droplet, once formed, will be relatively independent of the pump power, which determines the average exciton density throughout the sample, but does not affect the equilibrium chemical potential by much. The excitonic, or the insulating, nature of the droplets could be established by careful monitoring of evolution of the luminescence spectrum edge with temperature [1]. The Bose-Einstein condensation is a more subtle and difficult issue to address due to the reduced dimensionality [18]. It may manifest itself in interesting ways by its coupling to the photon field [19].

To summarize, we propose that a large droplet of a high density excitonic insulator can be stabilized in a suitably arranged quantum well structure where the electrons and holes are spatially separated. The argument given is quite general, and the actual dimensions of the quantum well structures, while affecting the density of the droplet, are not crucial to its formation. Such exciton droplets would be an example of the long sought-after high density Bose condensed phase of excitons, and allow one to examine the effects of coupling of a coherent electron-hole system with light, which may be itself coherent, as well as other interesting properties of the condensate.

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[14] These equations differ from the usual BCS gap equations [13] by factors of 2 at several places, because here we are explicitly calculating the energy of an electron-hole pair, rather than of a single electron participating in pairing.

[15] X. J. Zhu, P. B. Littlewood, M. S. Hybertsen, and T. M. Rice (unpublished).

[16] Not including the spin degeneracy, the densities are $1.3 \times 10^{11}/cm^2$ for $r_s = 2.66$ and $2.6 \times 10^{10}/cm^2$ for $r_s = 5.90$ for GaAs quantum wells.

[17] Taking into account the dynamic screening effects will lower the energy. It is not clear if $E^{pair}(r_s)$ has a minimum at the so-called metallic densities of $r_s \sim 2-4$ for $d = 0$. For $d \neq 0$, the presence of the Hartree term makes it unlikely for such a maximum density droplet phase to occur. For some nonvariational work in 3D, see A. P. Silin, Fiz. Tverd. Tela **19**, 134 (1977) [Sov. Phys. Solid State **19**, 77 (1977)]; R. Zimmermann, Phys. Status Solidi (b) **76**, 191 (1976).

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