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Bose-Einstein condensation of excitons in two dimensions

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Bose-Einstein condensation (BEC) is shown to exist for a noninteracting system in two dimensions, provided that the single-particle energy levels have a discrete spectrum below the continuum of extended states. This BEC, unlike that for three-dimensional Bose gases, possesses off-diagonal order only of finite range, and is thus not in violation of the Hohenberg theorem. The observation of such a BEC in the transport properties of excitons moving on the heterointerface of a type-II semiconductor superlattice is suggested, bearing in mind the fact that the random potential fluctuations would give rise to localized states on such surfaces.
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Recently, Bose-Einstein condensation (BEC) has been observed in some dilute alkali atomic gases.[1–5] This discovery is strongly dependent on the technology of trapping enough atoms in a very small region by laser cooling and magnetic trapping. The transition temperature for these alkali atomic gases is tremendously low ($\sim 2\ \mu$K). This is due to the heavy mass of these atoms and their low density. The population in the ground state of the alkali gases in general is very low, varying from 1000 (Ref. 1) to 10^6 (Ref. 3) as a consequence of the inevitable loss of atoms during the cooling and trapping processes.

If we turn to condensed matter physics, examples of the effects of BEC abound. The condensation of Cooper pairs gives rise to superconductivity. That of liquid helium leads to superfluidity. As a result of the much lower electron mass in the former and the much greater density in the latter, the critical temperatures for these cases are much higher, roughly from $T_c \sim 4$ K for helium to tens of degrees K for superconductors. However, the role of interaction in the former and the overlapping of the Cooper pair wave functions in the latter[6] render the role played by BEC less clear-cut.

The effect of BEC has also been measured in excitonic systems in bulk semiconductors[7] such as Cu_2O. The singlet paraexcitons rather than the triplet orthoexcitons are favored to form a Bose-Einstein condensate since the electron-hole interaction causes the former to have a lower ground state. In confined systems, the situation is rather different. The finite-size effect on an ideal Bose liquid was studied by Barber and Fisher.[8] Mei and Lee used the pseudopotential method to calculate the energy spectrum, free energy, and other thermodynamic functions of an interacting hard-sphere Bose gas in a thin film.[9] Most recently, suggestions and efforts were made to study the effect of BEC on the excitons in a quantum well.[10,11]

In exactly two dimensions (2D) it is well known that no BEC exists for free bosons. This situation is changed drastically, as we shall see, for noninteracting two-dimensional bosons with localized states.

Let us assume that a boson moving in two dimensions possesses a discrete spectrum below its continuous spectrum of energy levels, much like the case of potential wells. In an N-boson system, the number of particles populating the states in the entire continuous spectrum is given by summing over the corresponding Bose-Einstein distributions

$$N_{\text{ext. states}} = \sum_{\vec{k}} N_{\vec{k}} = \sum_{\vec{k}} \frac{1}{e^{\beta(\epsilon_{\vec{k}}-\mu)}-1}, \tag{1}$$

where $\epsilon_{\vec{k}} = \hbar^2 k^2/2m$ even though the wave function may not be a plane wave over the entire spatial region. Converting the sum in Eq. (1) into an integral involves the density-of-state factor $\rho(\epsilon_{\vec{k}})$. Thus we have

$$N_{\text{ext}} = \frac{V_{2D}}{(2\pi)^2}\int_0^\infty \frac{2\pi k dk}{e^{\beta(\epsilon_{\vec{k}}-\mu)}-1} = \int_0^\infty \frac{\rho(\epsilon)d\epsilon}{e^{\beta(\epsilon-\mu)}-1}. \tag{2}$$

If the chemical potential μ is allowed to approach zero from the negative side, the above integral would diverge because of the zero denominator at the lower integration limit. This is true as long as $\rho(\epsilon)$ approaches a constant as $\epsilon \to 0$, thereby giving rise to the familiar absence of BEC in 2D. Physically the divergence of N_{ext} as $\mu \to 0$ means the maximum population in the extended states is infinite. These states in the continuum could then accommodate as many particles as they come.

On the other hand, in the presence of an additional discrete spectrum consisting, without loss of generality, of energy levels ϵ_0 and ϵ_1, such that $\epsilon_0 < \epsilon_1 < 0$, the chemical potential μ can no longer be allowed to become zero since the physical requirement

$$N(\epsilon_0) = \frac{1}{e^{\beta(\epsilon_0-\mu)}-1} \geqslant 0 \tag{3}$$

imposes the condition that $\epsilon_0 - \mu \geqslant 0$ or $-\mu \geqslant -\epsilon_0 = |\epsilon_0|$, i.e., $(-\mu)_{min} = |\epsilon_0|$. The population of extended states, according to Eq. (2), now has to satisfy the inequality

$$N_{\text{ext}} = \frac{V_{2D}}{(2\pi)^2}\int_0^\infty \frac{2\pi k dk}{e^{\beta(\epsilon_{\vec{k}}-\mu)}-1} \leqslant \frac{V_{2D}}{(2\pi)^2}\int_0^\infty \frac{2\pi k dk}{e^{\beta(\epsilon_{\vec{k}}+|\epsilon_0|)}-1}. \tag{4}$$

The maximum of n_{ext} is

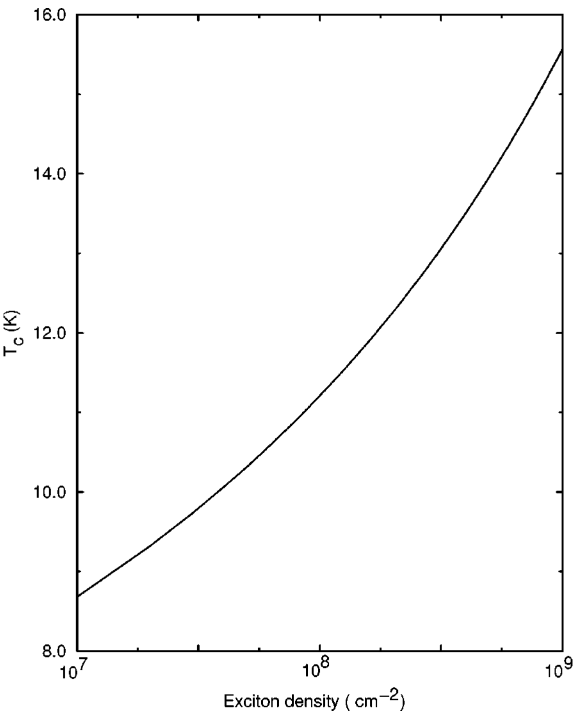

FIG. 1. The transition temperature T_c versus the density of excitons in type-II $(GaAs)_m/(AlAs)_n$ superlattices. The exciton is made of an electron with effective mass $m_0 = 0.066m_0$ and a heavy hole with effective mass $m_h = 0.50m_0$, where m_e is the bare electron mass. The ground energy $|\epsilon_0| = 6.8$ meV (Ref. 21).

$$(n_{\mathrm{ext}})_{max} = \frac{(N_{\mathrm{ext}})_{max}}{V_{2D}} = -\frac{1}{\lambda_T^2}\ln(1 - e^{-\beta|\epsilon_0|}), \tag{5}$$

where the integral in Eq. (4) has been evaluated exactly. Here $\lambda_T = (2\pi\hbar^2/mk_BT)^{1/2}$ is the thermal wavelength.

The existence of the upper limit $(n_{\mathrm{ext}})_{max}$ means that whenever the actual density n exceeds this limit, the extra amount $n - (n_{\mathrm{ext}})_{\mathrm{max}}$ will spill over to the discrete states, i.e.,

$$n - (n_{\mathrm{ext}})_{max} = n(\epsilon_0) + n(\epsilon_1). \tag{6}$$

Eq. (7) determines a critical temperature T_c in terms of the density n, the lowest energy level ϵ_0, and the boson mass m:

$$n = -\frac{1}{\lambda_c^2}(1 - e^{-\beta_c|\epsilon_0|}), \tag{7}$$

where $\beta_c = (k_BT_c)^{-1}$, $\lambda_c = \lambda(T_c)$. A plot of T_c versus the density n for given mass and ϵ_0 is shown in Fig. 1.

As T is decreased below T_c, n becomes greater than $(n_{\mathrm{ext}})_{max}$ and the extra particles $n - (n_{\mathrm{ext}})_{max}$ will be Bose condensed into localized states in macroscopic quantities. This is much like the familiar BEC for free bosons in 3D, except that the latter condensation is into a single $|\vec{k} = 0\rangle$ state that ranges coherently over the entire volume. Indeed, the single-particle density matrix $C(\vec{x}_1 - \vec{x}_2) \equiv \langle \hat{\psi}^\dagger(\vec{x}_1)\hat{\psi}(\vec{x}_2)\rangle$ can be obtained by expanding the field operator $\hat{\psi}(\vec{x}_1)$ into the complete set of eigenstates $\{\phi_i\} \equiv \{\phi_b;\phi_{\vec{k}}\}$ of the Hamiltonian of our present system. Therefore,

$$\begin{aligned}
C(\vec{x}_1,\vec{x}_2) &= \sum_i \sum_j \phi_j^*(\vec{x}_1)\phi_i(\vec{x}_2)\langle a_j^\dagger a_i\rangle_T \\
&= \frac{1}{V_{2D}}\sum_{\vec{k}} \sum_{\vec{k}'} e^{-i\vec{k}'\cdot\vec{x}_1} e^{i\vec{k}\cdot\vec{x}_2}\langle a_{\vec{k}'}^\dagger a_{\vec{k}}\rangle \\
&\quad + \sum_{b'} \sum_b \phi_{b'}^*(\vec{x}_1)\phi_b(\vec{x}_2)\langle a_{b'}^\dagger a_b\rangle \\
&= \sum_{\vec{k}} e^{-i\vec{k}\cdot(\vec{x}_1 - \vec{x}_2)}\langle n_{\vec{k}}\rangle_T + \sum_b \phi_b^*(\vec{x}_1)\phi_b(\vec{x}_2)\langle N_b\rangle_T \\
&= C_{\mathrm{ext}} + C_{local},
\end{aligned} \tag{8}$$

where the cross terms between the extended $|\vec{k}\rangle$ states and the bound $|b\rangle$ states make no contribution since $\langle a_{\vec{k}}^\dagger a_b\rangle = 0$. The first term in Eq. (8) arising from the extend states can be calculated exactly. We find for $x = |\vec{x}_1 - \vec{x}_2| \gg \lambda_T$,

$$C_{\mathrm{ext}}(\vec{x}_1,\vec{x}_2) = \frac{\sqrt{2\pi}}{\lambda_T^2}\frac{e^{-x/l}}{\sqrt{x/l}}, \tag{9}$$

where $l = (\lambda_T/\sqrt{4\pi})e^{(n_{\max}\lambda_T^2/2)}$, and $n_{\max} = -(1/\lambda_T^2)\ln(1 - e^{\beta|\epsilon_0|})$. This term, which is present even without BEC, decays exponentially on the length scale of l, the corresponding coherence length. The second contribution in Eq. (8) from the macroscopically populated localized or bound states in BEC depends obviously on the range of the individual localized wave functions. This $C_{local}(\vec{x}_1,\vec{x}_2)$ is nonvanishing only when both $\vec{x}_1$ and $\vec{x}_2$ are within the range of the individual localized states. Upon transition into the phase of BEC, the off-diagonal order characterized by $C(\vec{x}_1,\vec{x}_2)$ is thus seen to have only a finite range, of the order of the range of localization. This is in contrast to the BEC in 3D, which possesses off-diagonal–long-range order (ODLRO).[12] This also shows that the BEC in our system does not violate the Hohenberg theorem that no ODLRO can exist in 2D.[13]

Strictly speaking, the condensation is only into one single localized state, namely, the lowest state with energy ϵ_0. The population in the higher localized states would be given by the BE distribution function with the chemical potential μ set equal to the negative ϵ_0. However, the actual populations of the localized states may differ, depending on the nature of interaction between bosons. If the interaction upon close contact is repulsive or hard spherelike, the BE condensation of the bosons into the ϵ_0 state of finite spatial extent would eventually saturate and spill over to the next higher ϵ_1 state, etc. This would lead to successive saturation of the localized states one by one as the temperature dips below T_c, populating them macroscopically as a whole. The relative distribution among the localized states in each of which the particles are packed together would thus depend significantly on the interaction and the spatial range of these states. If the density n should become so large that $n - (n_{\mathrm{ext}})_{max}$ exceeded the total capacity of the localized states, the superfluous particles would feed back the extended states that must now be modified somewhat by the interaction. Yet the conclusion in Eq. (6) that the total population of the localized states becomes macroscopically large in the BEC phase should remain generally valid, in spite of the interaction.

One may work out the heat capacity and the various thermodynamic functions.[9,14] The most important characteristic of this BEC in 2D, however, is its transport properties. Upon the decrease of temperature across T_c of Eq. (7), a macroscopically large number of particles would precipitously fall into the localized states, which renders them much less mobile. A measurement of the outward diffusion current density (but not just the diffusivity) from the source region for neutral bosons, or of the electrical conductivity and magnetic susceptibility for charged bosons, should show telltale signs of the BEC. For instance, in the case where $\beta_c|\epsilon_0|\gg 1$, the BE distribution of the mobile particles becomes Boltzmann like for $T<T_c$. The electrical conductivity σ can then be approximated by the classical Drude model. Ignoring the particles condensed into localized states, we immediately obtain, by using Eq. (5) and assuming a mildly temperature-dependent mean free time τ, that

$$\sigma=\begin{cases} \dfrac{e^2\tau}{m}n, & T>T_c \\ \dfrac{e^2\tau}{2\pi\hbar^2}\dfrac{e^{-\beta|\epsilon_0|}}{\beta}, & T<T_c. \end{cases} \tag{10}$$

This attests to the sharp decrease in σ as T falls below T_C. Note the presence even in this Drude model of the quantum mechanical prefactor $T/\hbar^2$, in addition to the Boltzmann exponential factor that usually accompanies potential trapping. This prefactor is a distinguishing vestige from the bosonic coherence as expressed in the off-diagonal order of Eq. (8) and $(n_{\rm ext})_{max}$ of Eq. (5) that accompany phase transition into BEC.

The remaining question is where best to find a two-dimensional bosonic system with localized states. A good place to look is the heterointerfaces of type-II superlattices of semiconductor layers. For example, in a superlattice of alternate GaAs and AlAs layers, excitons are formed by the Coulomb attractive interaction of holes residing at the Γ point of the GaAs layers with the electrons residing at the X conduction-band edges of the AlAs layers.[15,16] These type-II excitons are thus indirect in both real and momentum space.[17] Each of these excitons straddles the heterointerface with the electron on one side and the hole on the other. The ground state pertaining to the internal structure of such an exciton has a binding energy somewhat less than $1/2^2$ Ry* (Ref. 14) owing to the special boundary condition that forbids the changing sides of electron and hole. These ground-state excitons are identical bosons that hug the interface and can move bodily on the surface. They constitute a perfect boson system in 2D as long as the exciton size is small compared to the spacing between them. This is in contrast to the excitons[18] moving within a quantum well of finite thickness,[10,11] which is only a quasi-two-dimensional system since very often the thickness is comparable to the size of the exciton. Furthermore, the unavoidable disorder or potential fluctuations[19] at the heterointerfaces inevitably give rise to localized states[20] that are of crucial importance to our considerations. Indeed, in a recent experiment by Gilliland *et al.*,[21] it was determined that the mobile excitonic states are about 6.8 ± 1.5 meV higher in energy than the distribution of localized states for a certain $(\mathrm{GaAs})_m/(\mathrm{AlAs})_n$ superlattice, which means in our considerations that $|\epsilon_0|$ in Eq. (7) is about 6.8 meV. A technique similar to that remarkable optical method developed by these workers whereby excitonic photoluminescence can be spatially and temporally resolved may provide a suitable means to observe the transport properties of the BEC and verify the relation (7) between the energy $|\epsilon_0|$, density n, and T_c in these two-dimensional bosonic systems. On the other hand, while a mobile exciton within a type-I quantum well[10] may be trapped by the donor dopants, it is unlikely that the trapped exciton can retain its internal structure intact. It would just become a different species of composite particles, not relevant to the consideration of BEC.

Generally speaking, if there are localized states in a Bose system of 3D, the BEC of the particles would again be into the localized states rather than the $|\vec{k}=0\rangle$ state, with off-diagonal order only of finite range. The essential difference is that there would always be localized states in a two-dimensional system with macroscopic disorder or potential fluctuations while the amount of disorder must exceed a certain threshold for localized states to appear in 3D.[20]

In the high-T_c cuprate superconductors, it is widely accepted that the Cooper pairs in the copper oxide layers play a crucial role. If we postulate the existence of such pairs even before the cuprate becomes superconducting, the random potential fluctuations in these layers would give rise to localized states for the bosonlike Cooper pairs (of small spatial extent). The BEC of these bosons in each layer may then exert a subtle influence on the behavior in both the normal and the superconducting states.[22,23]

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