Excitonic Instability and Electric-Field-Induced Phase Transition Towards a Two-Dimensional Exciton Condensate

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We present an InAs-GaSb–based system in which the electric-field tunability of its 2D energy gap implies a transition towards a thermodynamically stable excitonic condensed phase. Detailed calculations show a 3 meV BCS-like gap appearing in a second-order phase transition with electric field. We find this transition to be very sharp, solely due to exchange interaction, and, so, the exciton binding energy is greatly renormalized even at small condensate densities. This density gradually increases with external field, thus enabling the direct probe of the Bose-Einstein to BCS crossover. [S0031-9007(96)00671-0]

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The long search for a condensed phase of excitons has greatly expanded in recent years [1]. Although theoretically recognized three decades ago [2], experimental indications of the existence of such a phase emerged only recently. In Cu_2O bulk samples, The time evolution of the excitonic line shape [3] as well as macroscopic ballistic transport of excitons [4] are the most remarkable manifestations of the appearance of an exciton condensed phase. Indications of a two-dimensional (2D) condensate under strong magnetic field in semiconductor heterostructures were also reported [5,6].

Two major obstacles interfere in forming an exciton condensate. One is the recombination process which, apart from producing heat, can be faster ($\sim$1 ns) than the thermalization and condensation times. In Cu_2O, its unique crystal structure results in a dipole-forbidden recombination time of $\sim 10\ \mu$s. A usual way to increase the exciton lifetime in 2D structures is to introduce a wide barrier material between separated electron and hole materials [5]. This procedure, however, also reduces the electron-hole (*e-h*) Coulombic interaction, and so decreases T_c, the critical temperature for condensation. The second obstacle is the usually more favorable formation of an *e-h* droplet. Although that liquid phase may have a nonzero condensate order parameter, this is usually suppressed by the interactions leading to the formation of the droplet. It was suggested that these interactions can be reduced by strong magnetic fields [7], or in a sophisticated heterostructure design [8].

Both obstacles are immediately eliminated in a thermodynamic equilibrium condensed excitonic phase. Such is the state of a system with an excitonic instability [9,10], namely, a system with a semiconducting-like band structure, but with a single-particle energy gap which is smaller than the exciton binding energy. Excitons now form spontaneously, with no recombination. Compared to the unstable semiconducting ("vacuum") state, the excitonic state has negative energy, while a state of free *e-h* plasma has a positive energy. Therefore, at temperatures lower than the original energy gap, even if an *e-h* droplet forms, it must have a nonzero order parameter, and the excitonic phase exists even at high densities.

If, in addition to being unstable, the system allows external control over the original energy gap, then another important feature arises: The density of the exciton condensate can be externally controlled, and different density regimes can thus be probed in an otherwise identical system.

In this paper we present a realistic 2D system which exhibits an excitonic instability. Its band structure is very sensitive to externally applied electric fields. In particular, by application of field, one can continuously open an energy gap ranging from zero to a few meV. The negative (semimetallic) case is not discussed here.

We rigorously study the system using the Keldysh-BCS theory, described later. Starting from the large-gap state, we find an excitonic-condensate phase transition occurring exactly when the gap reaches the value of the exciton binding energy ε_0. Closing the gap further increases the condensate density n, up to $nd_0^2 \sim 4$ at zero energy gap. $d_0 = 2\hbar/\sqrt{2m\varepsilon_0}$ is the exciton Bohr diameter (m is the reduced effective mass). We use zero temperature and isotropic band structures of the underlying materials. However, in view of the instability origin of the condensed phase, low finite temperatures, warping, and imperfections should not change the results significantly.

A prototype of the studied heterosystem was described in Ref. [11]. Its energy profile is shown in Fig. 1. In this staggered-gap system, the valence band edge in the GaSb layer lies above the conduction band edge of the InAs layer. Because of the strong mismatch between valence and conduction states, a thin (i.e., 2–4 atomic layers) AlSb barrier is enough to suppress any *e-h* coupling between the active materials [12], and we have a regular semimetallic parabolic band structure. Such a thin barrier, however, has a negligible effect on the interlayer coulomb interaction. We designate this case as case (I). Without an

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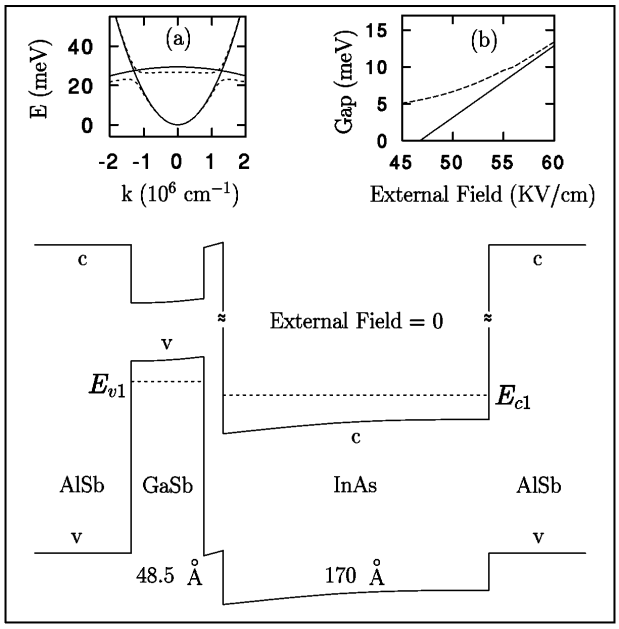

FIG. 1. Relevant band edges of the studied heterostructure. v, c denote valence and conduction band edges, respectively. E_{v1}, E_{c1} are the first confined levels. Layer widths correspond to 8 monolayers GaSb and 28 monolayers InAs. Inset (a) shows the in-plane spectrum of the carriers in the structure at zero electric field (solid lines). Dashed lines: Spectrum of coupled particles in a structure without the narrow AlSb barrier. Inset (b) shows the electric-field-dependence of the energy gap $E_{c1} - E_{v1}$ for both cases.

AlSb interlayer, there exists quantum mechanical coupling between the two layers [13], and an indirect-gap spectrum is formed [11]. This is case (II).

In both cases, small electric fields applied across the heterostructure greatly change its band structure. In case (I) the band overlap decreases with field, until a zero gap is reached, beyond which a semiconductor energy gap opens. In case (II), the main effect of the field is to shift the band extrema towards smaller wave numbers. An anticrossing energy gap of value 2–5 meV is now present at any field. However, it is possible to reduce that value by means of a very narrow AlSb barrier, or any other degradation of the GaSb-InAs interface. These results are summarized in insets (a) and (b) of Fig. 1. It is important to note that the total charge in the system can be independently controlled by means of a double-gated structure with in-plane leads [11], so we are allowed to consider a neutral sample even if the structure is originally unintentionally doped.

An excitonic instability happens in cases (I), (II) when the gap is smaller than the exciton binding energy ε_0. In case (II) the particles at the band edges are of mixed type, so ε_0 is much reduced. We therefore treat in this work only case (I). The formal way to treat the instability is by renormalization of the band structure [10,14,15]. This amounts to a BCS-like procedure, where the electrons and holes play the role of opposite-spin particles in the regular BCS theory. The outcome of the theory can be summarized in terms of $\Delta(k)$, the BCS gap function, and n, the condensate density. When $nd_0^2 \ll 1$, n is actually the exciton density, and the condensed phase is a Bose-Einstein condensate (BEC) of the Kosterlitz-Thouless type. In the opposite limit, n designates the density of "Cooper pairs," which are zero momentum e-h complexes in the Cooper-pairing sense. The transition between the two regimes is believed to be continuous [15,16]. We suggest the present system as an experimental platform to examine this BEC to BCS crossover.

To formulate the theory we start with the e-h Hamiltonian

$$H = \sum_{\mathbf{k}} (\varepsilon_{\mathbf{k}}^{e} a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} + \varepsilon_{\mathbf{k}}^{h} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}}) + \frac{1}{2D^2} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} (V_q^{ee} a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}'}^{\dagger} a_{\mathbf{k}'+\mathbf{q}} a_{\mathbf{k}-\mathbf{q}} + V_q^{hh} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}'}^{\dagger} b_{\mathbf{k}'+\mathbf{q}} b_{\mathbf{k}-\mathbf{q}} - 2V_q^{eh} a_{\mathbf{k}}^{\dagger} b_{\mathbf{k}'}^{\dagger} b_{\mathbf{k}'+\mathbf{q}} a_{\mathbf{k}-\mathbf{q}}), \tag{1}$$

where $\varepsilon_{\mathbf{k}}^{e,h}$ are the electron and hole energies (before renormalization), measured from their respective band edges, and $V_q^{\alpha\beta} = (2\pi e^2/\epsilon q) F^{\alpha\beta}(q)$. The structure factors $F^{\alpha\beta}(q)$ are found to be

$$F^{\alpha\alpha}(q) = \frac{8\pi^2}{\zeta_\alpha(4\pi^2 + \zeta_\alpha^2)} \times \left[1 + \frac{3\zeta_a^2}{8\pi^2} - \frac{4\pi^2}{(4\pi^2 + \zeta_\alpha^2)\zeta_\alpha}(1 - e^{-\zeta_\alpha})\right],$$

$$F^{eh}(q) = \frac{16\pi^4}{\zeta_e \zeta_h} \frac{(1 - e^{-\zeta_e})(1 - e^{-\zeta_h})}{(\zeta_e^2 + 4\pi^2)(\zeta_h^2 + 4\pi^2)},$$

where $\zeta_{e,h} = qL_{e,h}$, $L_{e,h}$ being the widths of the wells. It is worth mentioning that the structure factors reduce the binding energy, as well as the BCS gap, by more than a factor of three, as compared to the truly 2D case.

As usual [10,14,15], we proceed by making a Bogoliubov transformation of H, which results with a numerical (not operator) term in the Hamiltonian. Minimizing this term with respect to the transformation constant leads to the generalized BCS equations

$$\xi_{\mathbf{k}} = \varepsilon_{\mathbf{k}} + E_g - \int V_{\mathbf{k}-\mathbf{k}'}^{pp} \left(1 - \frac{\xi_{\mathbf{k}'}}{E_{\mathbf{k}'}}\right) \frac{d^2k'}{(2\pi)^2}, \tag{2a}$$

$$\Delta_{\mathbf{k}} = \int V_{\mathbf{k}-\mathbf{k}'}^{eh} \frac{\Delta_{\mathbf{k}'}}{E_{\mathbf{k}'}} \frac{d^2k'}{(2\pi)^2}, \tag{2b}$$

$$E_{\mathbf{k}} = (\xi_{\mathbf{k}}^2 + \Delta_{\mathbf{k}}^2)^{1/2}. \tag{2c}$$

Here $\varepsilon_{\mathbf{k}} = \varepsilon_{\mathbf{k}}^{e} + \varepsilon_{\mathbf{k}}^{h}$, $V^{pp} = \frac{1}{2}(V^{ee} + V^{hh})$, and E_g is the energy gap.

The integral term in Eq. (2a) results from the particle-particle Fock energy. When this term is set to zero, the system (2) becomes the regular BCS gap equation. We show later that the Fock term is uniquely responsible for the sharpness of the phase transition.

The outcomes of the self-consistent equations (2) are the BCS gap function $\Delta_{\mathbf{k}}$, which determines the renormalized spectrum $E_{\mathbf{k}}$, and the condensate density [14]

$$n = \frac{1}{2\pi^2}\int\left(1 - \frac{\xi_{\mathbf{k}}}{E_{\mathbf{k}}}\right)d^2k\,, \tag{3}$$

which includes here spin degrees of freedom.

The onset of the condensed state is found by solving Eqs. (2) in the first approximation in $\psi_{\mathbf{k}} \equiv \Delta_{\mathbf{k}}/E_{\mathbf{k}}$, where they become

$$\varepsilon_{\mathbf{k}}\psi_{\mathbf{k}} - \int V^{eh}_{|\mathbf{k}-\mathbf{k}'|}\psi_{\mathbf{k}'}\frac{d^2k'}{(2\pi)^2} = -E_g\psi_{\mathbf{k}}\,. \tag{4}$$

This is exactly the Schrödinger equation for the excitonic problem. It is therefore concluded that the phase transition occurs precisely at $E_g = \varepsilon_0$, regardless of the shape of the spectrum, and the finite width of the wells.

We proceed by solving Eqs. (2) for any E_g (i.e., any applied voltage). Results for n and Δ are shown in Figs. 2 and 3. Starting from large gaps we reach a second order phase transition when $E_g = \varepsilon_0$. Below ε_0, the condensate density gradually grows towards its value of $nd_0^2 = 3.8$ at $E_g = 0$. The results near $E_g = 0$ are of less significance, due to finite temperature effects. Screening is also expected to become important for $nd_0^2 \gg 1$. However, we see that for $E_g > 0.5$ meV, where low finite temperatures are irrelevant, all density regimes are present.

The same phase transition is also seen by considering the gap function Δ. Unlike the BCS behavior, $\Delta(k)$ is a nonmonotonic function of the wave number k. This is solely due to the exchange interaction term, which acts effectively as negative kinetic energy. When exchange is not neglected, ξ_k assumes negative values at small wave numbers, and the maximum of $\Delta(k)$ occurs roughly when ξ_k crosses zero.

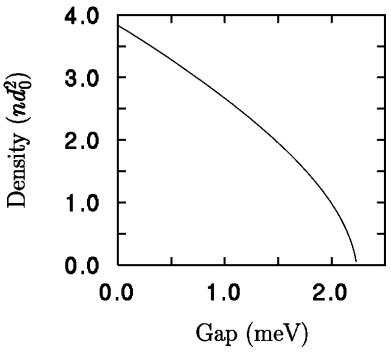

FIG. 2. Density of the condensate as a function of the original (not renormalized) energy gap. This gap scales linearly with external field.

The maximum value of $\Delta(k)$, $\Delta_{\max}$, is a measure of the strength of the condensate. Its value, about 3 meV, indicates that the condensate should be readily observed at helium temperatures in fair-mobility samples. The Fock term in Eqs. (2) is again responsible for the nonmonotonic dependence of $\Delta_{\max}$ on E_g. It can be understood if negative values of ξ_k are considered as corresponding to single-particle semimetallic spectrum. Then, reducing E_g is equivalent to increasing the semimetallic overlap, which is known [9] to reduce Δ.

The most obvious way to experimentally identify the formation of the condensate is to measure the dependence of the optical absorbtion edge on electric field. This can be done directly or by measuring the typical temperature for the temperature-activated conductivity of the system. Starting from a wide-gap structure, the edge, which is at $E_{\min}$, the minimum value of E_k, equals E_g, with a separated excitonic peak at $E_g - \varepsilon_0$. Closing the gap shifts these signals to lower energies, and diminishes the exciton peak, until it totally vanishes when $E_g = \varepsilon_0$. Beyond this point, the absorbtion edge dramatically increases with decreasing field, and that is identified with the formation of the condensate. The fact that $E_{\min} \gg E_g$ while the excitons are still well separated, $nd_0^2 \ll 1$, means that the exciton binding energy is greatly enhanced when the excitons form a BEC. This renormalization of ε_0 is attributed to the interaction between excitons, which is repulsive, and comes about through the particle-particle exchange term. Although this interaction can be small

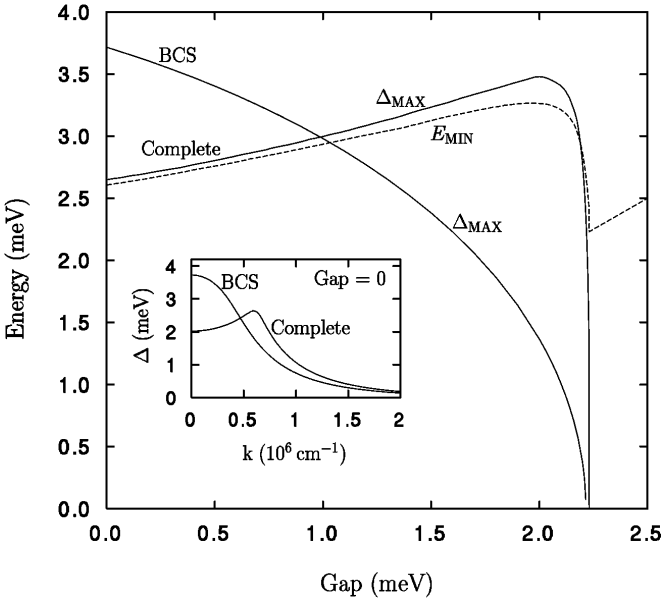

FIG. 3. Dependence of the condensate's parameters on the original gap. Solid lines: Maximum of Δ_k. Dashed lines: Minimum of E_k (i.e., the absorbtion edge). The inset shows $\Delta(k)$ for zero gap. "Complete" and "BCS" correspond, respectively, to calculations including, or excluding, the Fock term in Eq. (2a).

for small densities, it greatly affects the binding energy in a ground-state condensate, where the kinetic energy of each particle is zero. The effect is emphasized by the fact that $V^{pp}_{\mathbf{kk}'} \gg V^{eh}_{\mathbf{kk}'}$, the latter which governs the internal binding energy of the exciton. In contrast, if the exchange term is not included in the BCS equations, then the exciton binding energy (and $E_{\min}$) remains constant until $nd_0^2 \sim 1$.

The results provide a strong motivation to study the proposed material in the goal of achieving a strong, well-defined, stable, condensed phase of excitons. Unlike other proposed or experimentally studied structures, here we present a condensate which is inherent to the structure, and constitutes its equilibrium ground state even at temperatures as high as 0.5 meV. The density of the condensate can be easily changed here by means of electric field. Everything else being unchanged, the BEC to BCS crossover can thus be readily studied. Two previous works concerned excitonic phases in InAs-GaSb related structures. In [6] experiments were done in semimetallic conditions under strong magnetic fields. In [17] single-exciton binding energies were calculated for different well widths. Such calculations cannot account for the condensate even at very low densities where, as we have shown, many-body effects are of extreme importance. The effect of electric field on the condensate in these staggered-gap structures, and the externally induced phase transition, is recognized here for the first time.

Delicate questions concern the problem of whether the exciton condensate is superfluid. As is well known, macroscopic condensation in the ground state is not sufficient for superfluidity. Degeneracy of the quantum mechanical phase is also necessary. Indeed, It was argued [10,18] that the exciton condensate cannot be superfluid. The arguments are based on the suppression of superfluidity by interband transitions. Unanswered questions remain about the superfluid behavior in the limit when the matrix elements for those transitions are very small. Experiments in Cu_2O exhibit macroscopic ballistic transport of excitons [4], which can be interpreted as the result of superfluidity. Lozovik and Yudson [19], and Shevchenko [20] showed that if the electrons and holes are separated, the condensate can become superfluid (in that case it actually becomes a double-layer "capacitor superconductor"). Shevchenko recently presented a phase diagram for that case [21]. In the presently studied structure, electrons and holes are separated in type (I) structures. In type (II) structures the low-energy particles are of mixed type, and are spread across the double layer. Based on existing knowledge, one expects to experimentally find different superfluid behavior between the two structures, and so base the ideas behind the suppression of superfluidity on firm grounds.

In conclusion, we presented a prototype system which we believe should exhibit a condensed phase of excitons. This phase is the thermodynamic equilibrium state of the system. It is therefore expected to be robust to finite temperatures and imperfections. The parameters of the condensate are easily controllable by virtue of applied electric field. We thus propose the material as a new platform for studying the condensed phases of excitons.

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