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.

The long search for a condensed phase of excitons has greatly expanded in recent years [1]. Although theoretically recognised three decades ago [2], experimental indications of the existence of such a phase emerged only recently. In Cu$_2$O 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 [5-8].

Two major obstacles interfere in forming an exciton condensate. One is the short recombination lifetime ($\sim$ 1 ns) of photo-excited direct excitons. This time can be shorter than the time free excitons need to thermalize and condense. The recombination rate also produces enough heat to possibly destroy the condensate. In Cu$_2$O, 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 non-zero condensate order parameter, this is usually suppressed
by the interactions leading to the formation of the droplet. It was suggested [7] that a strong
magnetic field should reduce the interaction between excitons, thus favoring their quantum
condensation. Sophisticated heterostructure design has also been proposed [8] to overcome
the droplet formation.

Both of this obstacles are immediately eliminated if the thermodynamic equilibrium state
of the system is a condensed excitonic phase. Such is the case in 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, and the recombination lifetime is infinite. 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 non-zero 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.
The specific structure we propose can be readily fabricated by existing epitaxial techniques.
Its 2D band structure is very sensitive to externally applied electric fields. In particular, by
applying electric 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 properties of the system using the Keldysh-BCS theory, de-
scribed later. Starting from the large-gap state, we find an excitonic-condensate phase transition which occurs exactly when the gap reaches the value of the exciton binding energy \( \varepsilon_0 \). Closing the gap further results in an increase of the condensate density \( n \), up to a density of \( nd_0^2 \sim 4 \) at zero energy gap. \( d_0 = 2\hbar/\sqrt{2m\varepsilon_0} \) is twice the exciton Bohr radius (\( m \) is the reduced effective mass). In the theory we use zero temperature, and isotropic band structures of the underlying materials. However, in view of the instability-origin of the condensed phase, finite temperature, warping, and imperfections should not change the results significantly (as long as the temperature is well below the energy of the original gap).

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. Due to 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 AlSb interlayer, there exists quantum mechanical coupling between the two layers [13]. The band structure of the system is then composed of 2 W-shaped bands [11], and the particles at the band edges are of mixed type. 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. This gap now increases with applied field at a rate of 1 meVcm/KV. In case (II), the band structure is an indirect-gap-semiconductor one, with the main effect of the field being the shift of the band extrema towards larger wavenumbers. In this case an anticrossing energy gap of value 2-5 meV is present at any field. However, it is possible to reduce the value of the gap 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 $\varepsilon_0$. In case (II) the particles at the band edges are of mixed type, so $\varepsilon_0$ is much reduced. We therefore treat in this work only case (I), where no coupling is allowed. 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, which describes the band renormalization, 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 [13,16]. 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

$$H = \sum_k (\varepsilon^e_k a_k^\dagger a_k + \varepsilon^h_k b_k^\dagger b_k) +$$

$$\frac{1}{2D^2} \sum_{k,k',q} \left( V^e_{q} a_k^\dagger a_{k'}^\dagger a_{k'+q} a_{k-q} + V^h_{q} b_k^\dagger b_{k'}^\dagger b_{k'+q} b_{k-q} - 2V^{eh}_{q} a_k^\dagger b_{k'}^\dagger a_{k'+q} b_{k-q} \right)$$

where $\varepsilon^e,h_k$ are the electron and hole energies (before renormalization), measured from their respective band edges, $V^\alpha_\beta = 2\pi e^2/(\epsilon q) F^\alpha\beta(q)$, $\epsilon$ the dielectric constant, and $D^2$ is the sample area. $a_k$ and $b_k$ are electron and hole annihilation operators. The structure factors $F^\alpha\beta(q)$ are found to be:

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

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

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:

$$
\xi_k = \epsilon_k + E_g - \int V_{pp}^{k-k'} \left( 1 - \frac{\xi_{k'}}{E_{k'}} \right) \frac{d^2k'}{(2\pi)^2} \quad (3a)
$$

$$
\Delta_k = \int V_{eh}^{k-k'} \frac{\Delta_{k'}}{E_{k'}} \frac{d^2k'}{(2\pi)^2} \quad (3b)
$$

$$
E_k = \left( \xi_k^2 + \Delta_k^2 \right)^{1/2}. \quad (3c)
$$

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

The integral term in Eq. (3a) results from the particle-particle Fock energy. When this term is set to zero, the system (3) becomes the regular BCS gap equation. We show later that the Fock term qualitatively alters the results. In particular, due to this term, the transition to the excitonic phase is very sharp.

The outcome of the self-consistent equations (3) are the BCS gap function $\Delta_k$, which determines the renormalized spectrum $E_k$, and the condensate density \[ n = \frac{1}{2\pi^2} \int \left( 1 - \frac{\xi_k}{E_k} \right) \frac{d^2k}{(2\pi)^2} \quad (4) \]
which includes here spin degrees of freedom. Eqs. (3) can be solved analytically in the limits of large and small densities [10,15]. The onset of the condensed state is found by solving Eqs. (3) in the first approximation in $\psi_k \equiv \frac{\Delta_k}{E_k}$, where they become:

$$
\epsilon_k \psi_k - \int V_{eh}^{k-k'} \psi_{k'} \frac{d^2k'}{(2\pi)^2} = -E_g \psi_k. \quad (5)
$$

This is exactly the schrödinger equation for the excitonic problem. It is therefore concluded that the condensed state starts to form precisely when the gap $E_g$ equals the exciton binding energy, regardless of the shape of the spectrum, and the finite width of the wells.

To solve the problem completely, we define $x_k \equiv \tan \varphi_k \equiv \frac{\xi_k}{\Delta_k}$ to obtain a single equation
By solving Eq. (6) we analyze the excitonic condensate as a function of the original energy gap of the unstable-spectrum.

Results for \( n \) and \( \Delta \) are shown in Figs. 2 and 3. The energy gap is easily controlled experimentally by means of a perpendicularly applied voltage. Starting from large gaps we reach a second order phase transition when \( E_g = \varepsilon_0 \). Below \( \varepsilon_0 \), the condensate density grows gradually 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 the fact that small deviation from \( T = 0 \) would change the picture. Screening effects are 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 should not have an effect, all density regimes are present.

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

The maximum value of \( \Delta(k) \), \( \Delta_{\text{max}} \), 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. 3 is again responsible for the nonmonotonic dependence of \( \Delta_{\text{max}} \) on \( E_g \). It can be understood if negative values of \( \xi_k \) 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 \( \Delta \).

The most obvious way to experimentally identify the formation of the condensate is to measure the dependence of the optical absorption 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 \( E_{\text{min}} \), the minimum value of \( E_k \), is located at an energy \( E_g \), with a separated excitonic peak at \( E_g - \varepsilon_0 \). Closing
the gap by application of field shifts these signals to lower energies, and diminish the exciton peak, until it totally vanishes when $E_g = \varepsilon_0$. Beyond this point, the absorption edge steeply increases with decreasing field, and that is identified with the formation of the condensate.

The fact that $E_{\min}$ dramatically increases for $E_g < \varepsilon_0$, 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 $\varepsilon_0$ 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 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 $V_{kk'}^{pp} \gg V_{kk'}^{hh}$, 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 $E_{\min}$) remain 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$_2$O 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 results from an instability of the small-gap band structure, and 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, and of the underlying band structure, are easily controllable by virtue of applied electric field. We thus propose the material as a new platform for studying the condensed phase of excitons.

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FIGURE CAPTIONS

Fig. 1. Relevant band edges of the studied heterostructure. $v$, $c$ denote valence and conduction band edges. $E_{v1}$, $E_{c1}$ 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.

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

Fig. 3. Dependence of the condensate’s parameters on the original gap. Solid lines: maximum of $\Delta_k$. Dashed lines: Minimum of $E_k$ (i.e., the absorption edge). The inset shows $\Delta(k)$ for zero gap. ”Complete” and ”BCS” correspond, respectively, to calculations including, or excluding, the Fock term in Eq. (3a).
Density ($n d_0^2$) vs. Gap (meV)
