Excitonic Josephson effect in double-layer graphene junctions

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We show that double-layer graphene (DLG), where an external potential induces a charge-imbalance between \textit{n}- and \textit{p}-type layers, is a promising candidate to realize an exciton condensate in equilibrium. To prove this phenomenon experimentally, we suggest to couple two DLG systems, separated by a thin insulating barrier, and measure the excitonic Josephson effect. For this purpose we calculate the ac and dc Josephson currents induced by tunneling excitons and show that the former only occurs when the gate potentials of the DLG systems differ, irrespective of the phase relationship of their excitonic order parameters. A dc Josephson current develops if a finite order-parameter phase difference exists between two coupled DLG systems with identical gate potentials.

The search for the long ago predicted excitonic insulator (EI) state has recently stimulated a lot of experimental work, e.g., on pressure sensitive rare-earth chalcogenides, transition metal dichalcogenides or tantalum chalcogenides\textsuperscript{1–5}. Theoretically the excitonic instability is expected to happen, when semimetals with very small band overlap or semiconductors with very small band gap are cooled to very low temperatures\textsuperscript{6,7}. To date there exists no free of doubt realisation of the EI, however, and even the applicability of the original EI scenario to the above material classes is a controversial issue\textsuperscript{5,8–10}. There are serious arguments why the EI in these bulk materials, if present at all, resembles rather a charge-density-wave state than a “true” superfluid exciton condensate exhibiting off-diagonal long-range order\textsuperscript{11,12}.

On these grounds a non-ambiguous experimental proof of a macroscopic phase coherent exciton condensate would be highly desirable. We suggest that measuring an (excitonic) Josephson-type effect might be invaluable in this respect. Two-layer systems of spatially separated electrons and holes that feature an attractive interlayer electron-hole coupling are particularly suitable for such an experiment. Here a condensate of excitons might occur when the tunneling between the layers is negligible, but the corresponding Coulomb interaction is not\textsuperscript{13}. Double-layer systems thereby inhibit the obstacles coming from interband transitions or the coupling to phonons, which inevitably occur in bulk materials and prevent a possible exciton condensation by destroying the $U(1)$ symmetry\textsuperscript{12,14,15}. It is also advantageous that (exciton) tunneling effects are experimentally well-accessible in double-layer systems.

Graphene-based double-layers (separated by an adequate dielectric, e.g., hexagonal boron-nitride) show great promise for realizing a corresponding setup, see Fig. 1. For double-layer graphene (DLG), a gate-bias across the layers creates a charge imbalance, whereupon the attractive Coulomb interaction between the excess electrons and holes on opposite layers raises the possibility of exciton formation. While a fine tuning of the band gap can be achieved by the external potential, the recombination of electrons and holes can be fully suppressed by the dielectric\textsuperscript{16}. Then, in the weak coupling regime, exciton condensation is triggered by a Cooper-type instability, where the particle-hole symmetry of the system ensures a perfect nesting between the electron Fermi surface and its hole counterpart in the \textit{n} and \textit{p}-type layers of a biased DLG system\textsuperscript{17–19}. Placing an (insulating) tun-
nel barrier between two such DLG systems, a (potential) Josephson current can be used to analyze whether or not an exciton condensate has been formed in each of the subsystems.

The tight-binding Hamiltonian we assume for a DLG subsystem $i$ ($i = l, r$, left or right) has the form

$$H_i = \sum_{k} \varepsilon_{k_{i}}^{\pm} a_{k_{i}}^{\dagger} a_{k_{i}} + \sum_{k} \varepsilon_{k_{i}}^{-} b_{k_{i}}^{\dagger} b_{k_{i}} + \frac{1}{N} \sum_{k,k',q} U_{k,k',q} a_{k+q}^{\dagger} a_{k-q} b_{k'}^{\dagger} b_{k'};$$

(1)

where $a_{k_{i}}^{(1)}$ and $b_{k_{i}}^{(1)}$ annihilate (create) electron quasiparticles in the $n$-layer and $p$-layer, respectively, with in-plane momenta $k$ and band dispersions

$$\varepsilon_{k_{i}}^{\pm} = \pm \gamma_0 \left[ 3 + 2 \cos \left( \frac{\sqrt{3} k_y}{2} \right) + 4 \cos \left( \frac{\sqrt{3} k_y}{2} \right) \cos \left( \frac{3 k_x}{2} \right) \right]^{\frac{1}{2}} \mp \mu_i.$$  

(2)

In the low carrier density regime, the effective band structure (2) should account for the effects of the intralayer Coulomb interaction. The corresponding particle transfer amplitude is parametrized by $\gamma_0$, which defines the unit of energy in what follows. The external potential $V_i$ determines the chemical potential: $\mu_i = V_i/2$. The interlayer Coulomb interaction leading to exciton formation is

$$U_{k,k',q} = \kappa \frac{e^{-d|q|}}{|q|} \cos \left( \frac{\Phi}{2} \right) \cos \left( \frac{\Phi'}{2} \right),$$

(3)

where $\kappa = g_s 2\pi/\epsilon$, $g_s = 2$, $\epsilon$ denotes the dielectric constant of the dielectric, and $N$ gives the total number of particles.\textsuperscript{20,21} Since electron-hole recombination is neglected in Eq. (1) all interlayer Coulomb interaction terms that do not preserve the number of electrons (or holes) in a single layer, e.g., $H_{li} \propto \sum_{q} a_{k_i+q}^{\dagger} a_{k_i} b_{k_i}^{\dagger} b_{k_i}$ or $H_{il} \propto \sum_{q} a_{k_i}^{\dagger} a_{k_i-q} b_{k_i}^{\dagger} b_{k_i}$. Note that the model (1) exhibits a $U(1)$ symmetry, which causes the phase of the EI order parameter to be undetermined.

A mean-field decoupling of the Coulomb interaction yields

$$H_i = \sum_{k} \varepsilon_{k_{i}}^{\pm} a_{k_{i}}^{\dagger} a_{k_{i}} + \sum_{k} \varepsilon_{k_{i}}^{-} b_{k_{i}}^{\dagger} b_{k_{i}} + \sum_{k} \Delta_{k_{i}}^{\pm} b_{k_{i}}^{\dagger} a_{k_{i}}^{\dagger} + \sum_{k} \Delta_{k_{i}}^{-} a_{k_{i}}^{\dagger} b_{k_{i}};$$

(4)

where we have introduced the EI order parameter function

$$\Delta_{k_{i}} = -\frac{\kappa}{N} \sum_{q} \frac{e^{-d|q|}}{|q|} \frac{1 + \cos(\Phi')}{2} (a_{k+q}^{\dagger} b_{k+q}),$$

(5)

where $\Phi = \Theta_{k_{i}+q} - \Theta_{k_{i}}$ and $\Phi' = \Theta_{k_{i}'} - \Theta_{k_{i}'}$ with $\Theta_{k_{i}} = \arctan(k_y/k_x)$. The phase of $\Delta_{k_{i}}$ determines the phase of the ground-state wave function.

We first solve the self-consistency equations for the EI order parameter at zero temperatures, assuming $\kappa = 7.0$ and $d = 2.5$. Figure 2 shows its modulus $|\Delta_{k_{i}}|$ within the first Brillouin zone. Apparently only electrons and holes near the Brillouin zone’s $K$- or $K'$-points are bound into excitons. These particles occupy the states closest to the Fermi energy and therefore will be most susceptible for electron-hole pairing. Since the external potential fixes the position of the Fermi energy, it determines the behavior of $\Delta_{k_{i}}$ as well. When $V_i$ is raised from zero, more and more states become available for an electron-hole pairing. As a result the EI order parameter function is finite in a larger region of the Brillouin zone and the total (momentum accumulated) order parameter increases. At $V_i \approx 2$ the EI order parameter attains its maximal value and starts to decreases if the gate voltage gets larger until the EI phase breaks down at about $V_i \approx 3$ where the Fermi energy coincides with the upper/lower edge of the conduction/valence band. The relatively sharp boundary confining the area of bound electron-hole pairs indicates a BCS-like pairing\textsuperscript{22}.

A Josephson effect occurs when two DLG systems, where quantum coherence is realized, will be coupled to each other\textsuperscript{23}. Assuming that the ground states in both subsystems are described by macroscopic wave functions, $\Psi_i = |\Psi_i| e^{i\phi_i}$ ($i = l, r$ index the left respectively right subsystem), which—for simplicity reasons—are assumed to be equal in modulus but may have a different phases. The current density that flows between the left and right subsystem is $j = \frac{e}{h} (\Psi_i \nabla \Psi_{r}^{*} - \Psi_{r} \nabla \Psi_{i}^{*})$ (we set both $\hbar = 1$ and the electron mass $m_e = 1$; $e$ denotes the electron charge). Obviously any finite phase difference $\Delta \phi = \phi_r - \phi_l$ prevents the current density from vanishing, i.e., a persistent tunnel current flows through the barrier.
The Hamiltonian of the coupled system is \( H = H_l + H_r + H_T \), with
\[
H_T = \sum_{k,p} T_{k,p} \left( a_{kl}^+ a_{pr} + b_{kl}^+ b_{pr} \right) + \sum_{k,p} T_{k,p}^* a_{pr}^+ a_{kli} + b_{pr}^+ b_{kli},
\]
(6)
describing the tunnel process. In the numerical work we approximate the tunnel matrix element \( T_{k,p} = \delta_{k,p} \).

Tunneling excitons cause an electron current in the n-layer. This current equals the one in the p-layer—which flows in the opposite direction however—in modulus. Below we adapt the approach outlined in Ref. 24 to the (coherent) exciton tunnel processes in DLG. For this we define the tunnel current as
\[
I(t) = -e\langle \dot{N}_a(t) \rangle = -ie \int_{-\infty}^{t} dt' \left\langle \left[ \dot{N}_a(t), H_T(t') \right] \right\rangle,
\]
with
\[
N_a = \sum_{p} a_{p}^+ a_{p},
\]
(8)
\[
\dot{N}_a = -i \sum_{k,p} T_{k,p} a_{p}^+ a_{kli} + i \sum_{k,p} T_{k,p}^* a_{kli}^+ a_{p},
\]
(9)
where \( \dot{N}_a(t) = e^{iH't} \dot{N}_a(t) e^{-iH't} \), \( H_T(t') = e^{iH't} H_T e^{-iH't} \), and \( H' = H_l + H_r \). Note that the chemical potential in the left and right DLG system may differ. We therefore use \( K_l = H_l - \mu_l \), \( K_r = H_r - \mu_r \), and \( K' = K_l + K_r \), and obtain
\[
H_T(t') = \sum_{k,p} T_{k,p} e^{-i\mu_T t'} a_{kli}^+(t') a_{p}(t')
\]
\[+ \sum_{k,p} T_{k,p}^* e^{-i\mu_T t'} b_{kli}^+(t') b_{p}(t'),
\]
\[+ \sum_{k,p} T_{k,p} e^{-i\mu_T t'} a_{kli}^+(t') a_{p}(t'),
\]
\[+ \sum_{k,p} T_{k,p}^* e^{-i\mu_T t'} b_{kli}^+(t') b_{p}(t'),
\]
(10)
and
\[
\dot{N}_a(t) = i \sum_{k,p} T_{k,p}^* e^{i\mu_T t'} a_{p}(t) a_{kli}(t)
\]
\[+ \sum_{k,p} T_{k,p} e^{-i\mu_T t'} a_{kli}^+(t) a_{p}(t),
\]
(11)
Then the time-dependence of operators in the interaction picture is given by \( a_{kli}(t) = e^{iK'T} a_{kli} e^{-iK'T} \). We furthermore introduce the applied junction voltage
\[
W = \mu_r - \mu_l,
\]
(12)
and the operators
\[
A(t) = \sum_{k,p} T_{k,p} a_{p}(t) a_{kli}(t),
\]
(13)
\[
B(t) = \sum_{k,p} T_{k,p}^* b_{p}(t) b_{kli}(t).
\]
(14)
With it the tunnel current takes the form
\[
I(t) = -(i)^2e \int_{-\infty}^{t} dt' \left\langle \left[ [A(t), A^+(t')] \right] e^{iW(t-t')}
\]
\[+ \left\langle \left[ [A(t), B(t')] \right] e^{iW(t-t')} + \left\langle [A(t), A(t')] \right] e^{iW(t-t')} + \left\langle [A(t), B(t')] \right] e^{iW(t-t')} - \left\langle [A^+(t), A^+(t')] \right] e^{-iW(t-t')} \right\rangle,
\]
(15)
Let us focus on the current due to tunneling excitons exclusively. This current is governed by the second and eighth term of Eq. (15):
\[
I_X(t) = 2e \text{Im} e^{iWt} X_{ret}(-W),
\]
(16)
containing the retarded Green function
\[
X_{ret}(W) = \int_{-\infty}^{t} dt e^{iW(t-t')} X_{ret}(t - t'),
\]
(17)
\[
X_{ret}(t-t') = -i \Theta(t-t') \left\langle \left[ A(t), B^+(t') \right] \right\rangle.
\]
(18)
At the end we factorize \( X_{ret}(W) \) into contributions stemming from the left and right subsystems. For these we

**FIG. 3.** (color online) Excitonic Josephson current in case of (i) two identical DLG subsystems (green triangles, \( V_l = 1.0 \) and \( V_r = 1.0 \)), (ii) equal left and right external potentials but different phases of the EJ order parameters (magenta squares, \( V_l = 1.0 \) and \( V_r = 1.0 \)), (iii) equal external potentials with EJ order parameters having the same phase in the left and right DLG systems (blue dashed line, \( V_l = 0.0 \) and \( V_r = 1.0 \)), and (iv) different phases and different external potentials in both DLG subsystems (red continuous line, \( V_l = 0.0 \) and \( V_r = 1.0 \)).
use the mean-field Green functions calculated with the Hamiltonian (4).

Figure 3 gives the time dependence of the tunnel current for four characteristic situations. We first consider the case that two identical DLG systems are coupled by a thin barrier, i.e., \( V_l = V_r, W = 0 \). Clearly, if both EI order parameters have the same phase, no current flows through the junction. A dc current arises when the left and the right systems differ in terms of the phase of the EI-order parameter: \( \Delta \phi = \phi_r - \phi_l = -\pi/2 \). An ac current appears, on the other hand, if a finite (constant) voltage is applied across the junction. Its frequency is \( 2W \), which coincides with the frequency of the Josephson current for coupled superconductors. An additional phase difference amplifies the tunnel-current amplitude and leads to a phase shift in the current.

We finally analyze the voltage-current characteristics as to its phase dependence. For this purpose, we switch off the gate voltage in the left DLG system (\( V_l = 0 \)) and fix its EI phase to \( \pi \). Now the gate voltage in the right DLG subsystem is tuned from \( V_r = 0 \) to \( V_r = 8 \) for two choices of the right EI’s phase: \( \phi_r = \pi \) and \( \phi_r = \pi/2 \). The corresponding results are displayed in Fig. 4. Most notably, at \( W = 0 \), a finite dc Josephson current only appears if \( \Delta \phi \neq 0 \). Otherwise \( \Delta \phi = 0 \) and \( \Delta \phi \neq 0 \) basically cause the same qualitative behavior. The amplitude increases with increasing voltage until it reaches its maximum at a junction voltage \( |W| = 1.0 \). This coincides with the point, \( V_r = 2.0 \), at which the EI order parameter in the right DLG system attains its largest value.

If the voltages grows further the amplitude of the exciting Josephson current vanishes rapidly. Unfortunately the current becomes extremely small just before the EI phase completely breaks down. Therefore, the Josephson tunnel current is not much suitable for a extremely precise determination of the EI phase boundary. Figure 4 corroborates that a finite phase difference yields a larger current amplitude.

To conclude, the setup proposed might be used to identify a condensed exciton phase in double-layer graphene (DLG), which is subjected to an external electric potential, by analyzing a Josephson-type effect in a DLG junction device. Provided the gate potentials of the DLG systems differ, an ac Josephson tunnel is observed irrespective of the phase relationship of the excitonic order parameters in both subsystems. If both DLG subsystems are exposed to the same gate voltages but there is a finite phase difference between their exciton order parameters, a dc current appears. Such a finite phase difference suggests a degeneracy of the ground state, i.e., a \( U(1) \) symmetry. This symmetry is closely related to off-diagonal long range order and only in this situation the exciting insulator represents a genuine exciton condensate\(^{12}\).

We like to emphasize that small leakage currents, which may arise in an actual experiment, are linked to interlayer hopping of electrons and holes or interlayer exchange terms due to the Coulomb interaction. These terms pin the phase \( \phi_i \) to a specific value and therefore destroy the \( U(1) \) symmetry\(^{14,15}\). Hence exciton condensation—in a strict sense—cannot occur and the excitonic insulator, if present, features a charge-density-wave state. Interlayer hopping and exchange terms like \( H_U \propto a^{\dagger}_{k+q} a_i a^{\dagger}_{k'-q} b_{k'i} \), moreover, enforce a finite \( \Delta_{ki} \) at all temperatures and therefore prevent a true phase transition\(^{15}\).

The present study, for sure, should be considered as first step towards a theoretical modelling of the excitonic Josephson effect. The mean-field approach used is certainly a crude approximation and any future (more detailed) analysis should rely on more elaborated methods that take into account fluctuation and correlation effects. Also the treatment of the tunnel junction could be improved, e.g., by using a more realistic (material specific) tunnel matrix element and including pair-breaking effects within the barrier. All this would improve the understanding of the fascinating exciton condensation phenomenon.

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1. B. Bucher, P. Steiner, and P. Wachter, Phys. Rev. Lett. 67, 2717 (1991).
2. P. Wachter, B. Bucher, and J. Malar, Phys. Rev. B 69, 094502 (2004).
