Density fluctuation effects on the exciton condensate in double-layer graphene

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Density fluctuation effects on the exciton condensate in double layer graphene

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We describe the robustness of an excitonic condensate in double layer graphene against layer density fluctuations and the associated charge inhomogeneity, and discuss the implications for observing the condensate in current experimental conditions. We solve the mean-field equations for a finite imbalance in the Fermi energies in each layer and utilize the results in two phenomenological models for inhomogeneity. We find that the stability of the excitonic condensate against density fluctuations is strongly dependent on the size of the excitonic gap, and that transport experiments (such as Coulomb drag) are promising methods for observing the condensate.

Excitonic condensates, where pairs of electrons and holes in a semiconductor-like system undergo a Bose Einstein condensation, have been predicted to occur in bilayer semiconductor heterostructures[1]. They have previously been observed experimentally under high magnetic fields in the quantum Hall regime[2,3]. Recent advances in fabricating double layer graphene (DLG), where a layer of dielectric is sandwiched between two graphene layers, have motivated theoretical predictions of the emergence of an excitonic condensate at zero magnetic field in these heterostructures[4,5] when one layer is electron-doped and the other is hole-doped. In DLG, the thick dielectric prevents any tunneling of carriers between the two layers, while the inter-layer (attractive electron–hole) Coulomb interaction drives the formation of excitonic condensate.

The estimate of the critical temperature T_c for excitonic condensation varies over a wide range, even within mean field theory, depending on the level of screening of the Coulomb interaction considered in the model. At high density, unscreened Coulomb interactions produce T_c approaching room temperature[5], while static screening leads to $T_c \lesssim 0.1\text{K}$[6,7]. Large wave vector scattering induced by short-ranged disorder further reduces T_c[8,9], while the inclusion of dynamic screening[10,11] and the full band structure[12] tend to favor the pairing. The controversial nature of the size of the excitonic gap is reinforced by the absence of any signature of the condensate in recent Coulomb drag experiments[13] on DLG. Since disorder scattering strongly suppresses T_c[8,9], the optimal regime to search for excitonic condensates in DLG is the relatively low density regime ($k_F d < 1$) where k_F is the Fermi wavevector and d the inter-layer separation. But this effectively low-density (i.e. low k_F) regime in graphene is susceptible to strong density fluctuation effects (the so-called electron-hole puddles[14]) arising from either extrinsic (Coulomb impurity-induced) or intrinsic (e.g. ripple-induced) charge inhomogeneity in the system. In the current work we explore the interesting and important question of the effect of inhomogeneous layer density fluctuations on the excitonic condensation, emphasizing that this is a distinct physical mechanism from impurity disorder,[8,9] where the latter mainly hinders the condensate formation through momentum-conservation-breaking scattering processes. The density fluctuations may be caused by any general scalar potential such as the Coulomb field of charged impurities or the effect of corrugations and ripples. But regardless of the origin of the potential, the key distinguishing feature of our work is that we allow for imperfect nesting of the Fermi surfaces due to the difference in chemical potential in the two layers which is likely to be the primary manifestation of disorder in these devices.

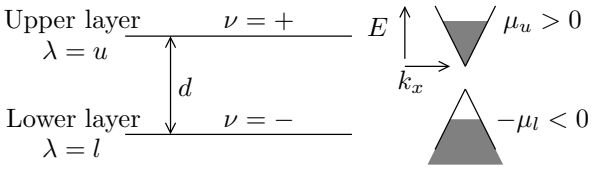

FIG. 1. Sketch of DLG doped such that the upper layer contains electrons and the lower layer contains holes with different densities.

It is well known that the charge landscape of graphene mounted on a substrate is inhomogenous[14–16] and that when the overall density is small, 'puddles' of electrons and holes are formed. Various mechanisms for the formation of this inhomogeneity have been proposed, including the presence of charged impurities in the environment[17,18] and ripples[19,20]. These density fluctuations can also be viewed as inhomogeneities in the local chemical potential[21]. The presence of the top and bottom gates, as well as the dielectric between the graphene layers, implies that such chemical potential fluctuations are ubiquitous in DLG samples. We study the effects of these fluctuations on the excitonic condensate within a local density approximation. Keeping in mind the wide variation of the excitonic gap in the clean case based on the theoretical approximation scheme, we look at the two extreme cases: (i) unscreened Coulomb interaction and (ii) Coulomb interaction with static screening, with a focus on common features obtained by scaling various energy scales by $\tilde{\Delta}_0$, which is the excitonic gap for the clean sample in either approximation.

Our main results are: (a) In spatially homogeneous DLG, with different elctron and hole Fermi energies, the

pairing profile Δ_k depends only on the average Fermi energy $\bar{\mu}$, and is independent of the difference $\delta\mu$ until $|\delta\mu| = 2\tilde{\Delta}_0$. Beyond this point, the pairing collapses for both the screened and the unscreened interaction. (b) For imbalanced Fermi surfaces, we find no evidence of a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) type state[22] with pairing at finite center of mass momentum $|\mathbf{Q}| = k_{Fu} - k_{Fl}$ in either the unscreened or the screened case. (c) We provide two different estimates of the effect of charge inhomogeneity on the excitonic condensate: (i) the average pairing gap, which is relevant for thermodynamic measurements and (ii) the fraction of the sample area supporting the condensate which is relevant for transport measurements. We discuss these quantities as a function of the average chemical potential and the width of density fluctuations for both screening models. Our results show that screening makes the excitonic condensates more vulnerable to charge fluctuations by virtue of the smaller excitonic gap and that transport measurements are more robust to these fluctuations than bulk thermodynamic measurements.

The large separation of the two graphene layers implies that the local chemical potential (and hence charge) fluctuations are independent of each other. For convenience, we will assume the top layer to be electron doped with local chemical potential μ_u and the lower layer to be hole doped with local chemical potential $-\mu_l$. These quantites may fluctuate spatially. The independent fluctuations of the two chemical potentials imply that the perfect nesting of Fermi surfaces is lost in these inhomogeneous materials even when the global average $\langle\mu\rangle$ is the same in each layer. Before going into the details of averaging over distributions of chemical potentials through a local density approximation, we examine the effect of unequal (but spatially homogeneous) chemical potentials in the two layers on excitonic properties.

The Hamiltonian for a single spin/valley species in monolayer graphene in layer $\lambda \in \{u, l\}$ is given by $H_{\lambda\nu} = \sum_{\mathbf{k}} (\varepsilon_{\mathbf{k}\nu} - \nu\mu_\lambda)\, c^\dagger_{\lambda\mathbf{k}\nu} c_{\lambda\mathbf{k}\nu}$ where $\nu \in \{+, -\}$ denotes the band and $\varepsilon_{\mathbf{k}\nu} = \nu v k$ is the single particle energy. We will set $\hbar = 1$ for the rest of this paper. For excitonic condensates in DLG, we are interested in the case where the average electron (hole) doping in the upper (lower) layer is large (i.e. $|\mu_\lambda| \gg \Delta_0$). Since exciton condensation is dominated by processes around the Fermi surfaces, we can exclude the filled (empty) valence (conduction) band in the upper (lower) layer from our model. The full Hamiltonian is then $H = H_{u+} + H_{l-} + V_{\rm eh}$ where $V_{\rm eh}$ is the inter-layer interaction between the electrons and holes. We neglect the intra-layer interaction since it amounts only to a small renormalization of the Fermi velocity in the layer[23,24]. We write the second-quantized operators for electrons in the upper layer as $a_{\mathbf{k}} = c_{u\mathbf{k}+}$ and for holes in the lower layer as $b_{\mathbf{k}} = c^\dagger_{l\mathbf{k}-}$. This particle-hole transformation allows us to make a direct connection with the BCS theory of superconductivity, albeit with a complicated and non-separable potential kernel. In this notation, the BCS pairing ansatz with

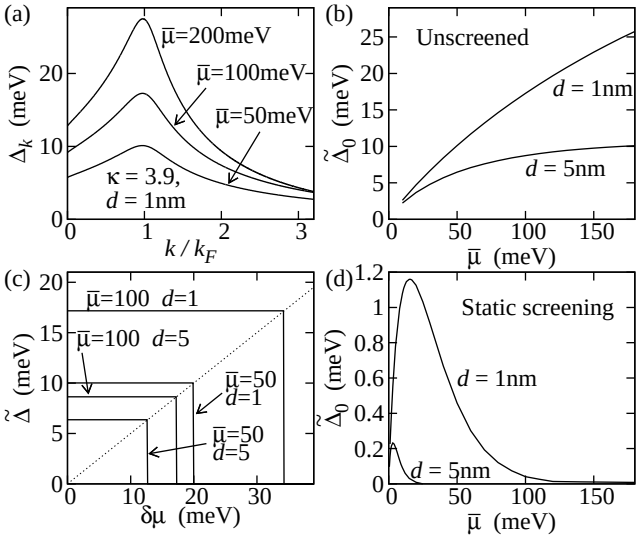

FIG. 2. (a) The pairing amplitude $\Delta_{\mathbf{k}}$ for unscreened interaction with $\delta\mu = 0$ and different $\bar{\mu}$. (b) The largest pairing amplitude $\tilde{\Delta}_0$ for the unscreened interaction at $\delta\mu = 0$. (c) The evolution of the largest pairing amplitude $\tilde{\Delta}$ with $\delta\mu$ for the unscreened interaction. The dotted line is $\delta\mu/2$ which serves as a guide to the eye illustrating the collapse of the excitonic pairing when $\delta\mu = 2\tilde{\Delta}_0$. (d) $\tilde{\Delta}_0$ for the statically screened interaction at $\delta\mu = 0$.

zero center of mass momentum for the interaction gives

$$V_{\rm eh} = \sum_{\mathbf{kq}} V(\mathbf{q}) f(\mathbf{k}+\mathbf{q}, \mathbf{k}) a^\dagger_{-\mathbf{k}-\mathbf{q}} b^\dagger_{\mathbf{k}+\mathbf{q}} b_{\mathbf{k}} a_{-\mathbf{k}} \qquad (1)$$

where $V(\mathbf{q})$ is the interaction potential and $f(\mathbf{k}, \mathbf{k}') = [1 + \cos(\theta_k - \theta_{k'})]/2$ is the chirality factor due to the graphene band wave functions. Using standard diagonalization techniques with a mean field in the excitonic channel, $\Delta_{\mathbf{k}} = \langle b_{\mathbf{k}} a_{-\mathbf{k}} \rangle$, we obtain two quasiparticle branches with spectrum $E_{\alpha,\beta} = \delta\mu/2 \pm \sqrt{(\hbar v k - \bar{\mu})^2 + \Delta^2_{\mathbf{k}}}$ where $\bar{\mu} = (\mu_u + \mu_l)/2$ and $\delta\mu = \mu_u - \mu_l$. The self-consistent gap equation is then given by

$$\Delta_{\mathbf{k}} = \sum_{\mathbf{k}'} V(\mathbf{k}' - \mathbf{k}) \frac{\Delta_{\mathbf{k}'} f(\mathbf{k}, \mathbf{k}') \left[n_\beta(k') - n_\alpha(k')\right]}{\sqrt{(\varepsilon_{\mathbf{k}'} - \bar{\mu})^2 + \Delta^2_{\mathbf{k}'}}} \qquad (2)$$

where the occupation numbers are $n_{\alpha(\beta)}(k) = \Theta(-E_{\alpha(\beta)}(k))$ at zero temperature. Equation (2) shows that $\bar{\mu}$ and $\delta\mu$ are the natural variables to analyze the system. We will first look at the system with $\delta\mu = 0$, where the two Fermi surfaces are perfectly nested.

Within a model of unscreened Coulomb interaction[4,5], which gives the most generous estimate for the excitonic gap, the inter-layer potential is $V(\mathbf{q}) \equiv V_b(q) e^{-qd} = 2\pi e^2 e^{-qd}/(\kappa q)$ where $V_b(q)$ is the bare 2D Coulomb interaction and κ is the dielectric constant of the environment[25] The momentum profile of Δ_k for $\delta\mu = 0$ is shown in Fig. 2(a) for a small inter-layer separation of 1nm and three different values of $\bar{\mu}$. Δ_k has a non-monotonic momentum dependence with a peak at $k = k_F$

whose height increases with the size of the Fermi surface. This peak height, $\tilde{\Delta}_0$, is shown in Fig. 2(b) as a function of $\bar{\mu}$ for two different values of the inter-layer separation, $d = 1, 5$nm. The profiles have a convex shape which flattens out at large values of $\bar{\mu}$. For $\delta\mu = 0$, the quasiparticle spectrum $E_\alpha = -E_\beta$ and the positive energy branch has a minimum $\tilde{\Delta}_0$ at $k = k_F$. Thus $\tilde{\Delta}_0$ can be identified as the true single particle spectral gap in the system.

In the opposite limit, we study the model of Coulomb interactions with static screening, which gives the lowest estimate of excitonic gaps at high density. In this case, the inter-layer potential is given by[7]

$$V_{\rm eh}(\mathbf{q}) = \frac{V_b(q)e^{-qd}}{1 - V_b(q)\left[\Pi_u + \Pi_l\right] + V_b(q)^2\Pi_u\Pi_l\left(1 - e^{-2qd}\right)}$$

where $\Pi_\lambda(q)$ is the polarization function in a single layer[26] which may be different in the two layers for mismatched Fermi surfaces. The momentum dependence of Δ_k takes a similar form to the unscreened case, with a peak at $k = k_F$, but, as shown in Fig. 2(d), the size of the peak $\tilde{\Delta}_0$ is several orders of magnitude smaller. Unlike the unscreened case, $\tilde{\Delta}_0$ shows a non-monotonic dependence on $\bar{\mu}$ with a peak occuring at a relatively small value of $\bar{\mu}$. The increase in $\tilde{\Delta}_0$ at small $\bar{\mu}$ is due to the increasing density of states at the Fermi level, but increasing the size of Fermi surface also makes the screening of the Coulomb interaction more efficient since the Thomas Fermi wave vector $q_{TF} \sim k_F$ in linearly dispersing graphene. At large densities, the increased screening overwhelms the increasing density of states and the excitonic gap decreases rapidly[6,7].

We now focus our attention on the effects of Fermi surface imbalance on the excitonic properties, using a pairing ansatz with zero center of mass momentum. We see that changing the sign of $\delta\mu$ is equivalent to changing $E_\alpha \to -E_\beta$ so a discussion of $\delta\mu > 0$ will suffice. As $\delta\mu$ increases, we find that the profile of Δ_k remains unchanged until $\delta\mu = 2\tilde{\Delta}_0$. For $\delta\mu > 2\tilde{\Delta}_0$, the pairing vanishes for all k leading to the normal state. This behaviour is common to both the unscreened and screened interaction model and is equivalent to the Clogston-Chandrasekhar[27] limit for loss of pairing in a superconductor in a Zeeman field. This is visible explicitly in Fig. 2(c), where the peak pairing amplitude $\tilde{\Delta}$ is plotted as a function of $\delta\mu$ for the unscreened interaction for $\bar{\mu} = 50$ and 100meV and $d = 1$ and 5nm. This behaviour can be understood from Eq. (2) where $\delta\mu$ appears only through the Fermi functions $n_{\alpha(\beta)}$. The profile for Δ_k is unchanged from the $\delta\mu = 0$ case so the occupation probabilities are unchanged if $|\delta\mu|/2 < \tilde{\Delta}_0$. Thus the profile for $\delta\mu = 0$ satisfies the gap equation for $\delta\mu < 2\tilde{\Delta}_0$. Beyond this value, the occupation probabilities are changed around the gap edge, where the gap edge singularities in the density of states ensure that the pairing is completely lost.

We have also looked at the possibility of electron-hole pairing with finite center of mass in the case of imbalanced Fermi surfaces, specifically for pairing with center of mass momentum $|Q| = |k_{Fu} - k_{Fl}|$, which has a rich history in the theory of superconductivity under the name "FFLO pairing"[22]. We have not found any evidence of FFLO type excitonic pairing in either the screened or the unscreened model. This is not surprising, since, even with the model of a constant local interaction, FFLO states are known to stabilize in only a narrow parameter regime in higher than 1D[28]. The momentum dependence of the Coulomb potential would further destabilize this fragile state. We safely predict the non-existence of any FFLO-type interlayer excitonic superfluidity in density-imbalanced bilayer systems.

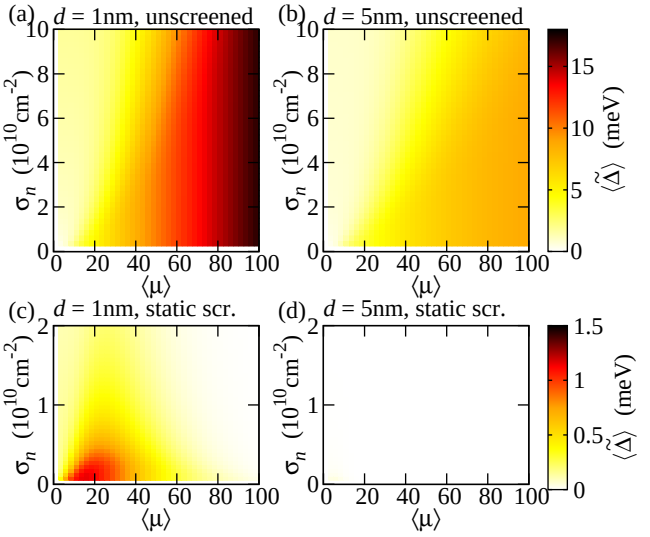

FIG. 3. The areally averaged excitonic gap, given by Eq. (3). The layer separation and screening model are labelled.

Having looked at the effects of Fermi surface imbalance on homogeneous states, we now connect these findings to excitonic condensates in actual inhomogeneous DLG samples. In the first instance, one could imagine a thermodynamic experimental probe which couples to an area of the sample which is larger than the size of the average fluctuation (e.g. measurement of a gap derived from specific heat). In this case, the measured excitonic gap might be best modeled as an average of the gap over the spatial area sampled. Since the correlation length of the density fluctuations is of the order of the system size, we can use a local density approximation to write

$$\langle\tilde{\Delta}\rangle = \iint d\mu_l\, d\mu_u\, P(\mu_u)P(\mu_l)\tilde{\Delta}(\mu_u, \mu_l) \qquad (3)$$

where $P(\mu_\lambda)$ is the probability distribution for the chemical potential in layer λ. Similar averaging procedures have been employed with great success[29]. It is well known that for monolayer graphene, the distribution of density fluctuations due to charge impurities is Gaussian[17,30], and hence

$$P(\mu) = \frac{2|\mu|}{\sqrt{2\pi^3}v^2\sigma_n}\exp\left[-\frac{(\mu|\mu| - \langle\mu\rangle|\langle\mu\rangle|)^2}{2\sigma_n^2 v^4\pi^2}\right] \qquad (4)$$

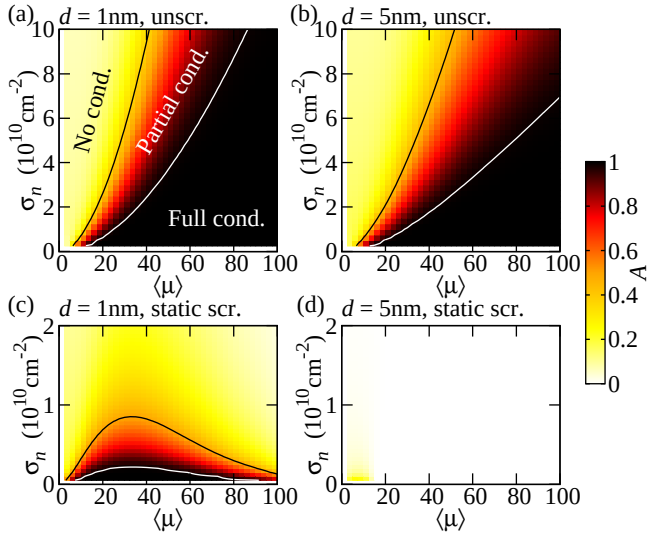

FIG. 4. The fraction of the area supporting the excitonic condensate in inhomogeneous DLG. The black contour denotes $A = 0.5$ and the white contour is $A = 1$.

where σ_n is the width of density fluctuations determined by impurity concentrations, $\langle\mu\rangle$ is the global chemical potential (which we assume to be the same for both layers and is set by external gating). It is known from theoretical work[17] that in the case of charged impurities, σ_n and the 2D impurity concentration are of the same order of magnitude. We assume that the inhomogeneities in the two layers are uncorrelated since if the density fluctuations are caused by ripples there is no reason to believe that these should be the same in the two layers, and the field due to charged impurities will be effectively screened from the more distant layer by the closer one. However, inter-layer correlations can be included by substituting a more sophisticated function instead of the product $P(\mu_u)P(\mu_l)$. Thus we have reduced the effects of inhomogeneity to one phenomenological parameter σ_n which takes the same value in both layers. Evaluating the integrals in Eq. (3) gives the color plots shown in Fig. 3. In the unscreened case for small inter-layer distance, the gap is large enough that the presence of density fluctuations does not substantially alter the gap. However, for static screening, the fluctuations are large enough to completely kill the exciton condensate for moderate inter-layer separation, and even for minimal separation at low density a vanishing amount of inhomogeneity is required if the gap is to persist.

A more definitive signature of excitonic condensates is the "superfluid" or perfectly inductive response of the system in drag transport measurements. The condensate and non-condensate regions form a network and percolation theory can be applied to determine the transport properties[31]. The strongly inhomogeneous samples most probably show glassy behaviour, but the nature of this glassy state requires a more sophisticated treatment of disorder. Although a full analysis of percolating clusters is beyond the scope of this paper, the fraction of area supporting the condensates can be related to percolation thresholds. The fractional area of the sample which supports the condensate is given by

$$A = \iint d\mu_u\, d\mu_l P(\mu_u)P(\mu_l)\Theta(\mu_u\mu_l)\Theta(2\tilde{\Delta}_0 - |\delta\mu|) \quad (5)$$

The first step function represents the fact that in the inhomogenous case it is possible for μ_u and μ_l to be in the same band. If this is the case then the condensate is not allowed. Figure 4 shows A for both the unscreened and statically screened interactions, and for the two values of the inter-layer separation shown previously. The black contour denotes $A = 0.5$, where half the area supports the condensate, and the white contour shows $A = 1$ meaning that the whole sample is excitonic. We see that in the unscreened case, the condensate should be stable at experimentally accessible values of density fluctuations, but in the statically screened case the fluctuations have to be smaller even than those currently available in hBN devices[30,32]. By comparing Figs. 4 and 3, we see that in samples with density fluctuations, the formation of the excitonic condensate is easier to detect in transport-style experiments than in probes that average over the bulk of the sample[33].

We also note that other authors[10,11] have considered the effect of dynamic screening on the exciton condensate without disorder. In the high density limit (where static screening gives a tiny gap due to the large size of the polarization function), dynamic screening gives a gap which is closer to the unscreened interaction than the static screening. Using the data presented in Ref. 10, we estimate that for physically realistic parameters, the dynamic screening theory predicts that the peak gap in the high density regime will be $\sim$ 0.1meV, indicating that the numerical effect of disorder is roughly similar to that of the static screening in the low density regime. Since our analysis shows that the nature of the screening has no impact on the effects of inhomogeneity, Figs. 3 and 4 show that a gap of this size is marginally observable. Therefore, just as the low density statically screened interaction may be observed in the cleanest systems, so should a gap generated by dynamic screening in the high density regime.

In summary, we have examined the impact of chemical potential fluctuations on the formation of an excitonic condensate in DLG. The origin of these charge fluctuations may be from ripples in the graphene surface, charged impurities in the environment of the graphene, or any other general spatially-varying scalar field. While the absolute value of the gap depends sensitively on the model of interaction used, the effects of Fermi surface imbalance are identical in the two cases. However, since the inhomogeneity can be controlled only on an absolute scale, we find that the size of the gap is the crucial factor which determines the robustness of the condensate against this form of disorder. If the gap is of the order of a few meV, then the level of fluctuations found with hBN

substrates should be sufficient to observe the condensate in Coulomb drag experiments. Finally, if static screening is important in the formation of excitonic condensates, there is an optimal Fermi energy (which is ~ 20meV for $d = 1$nm), where it would be easiest to see the condensate.

ACKNOWLEDGMENTS

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