Inter-layer excitonic superfluidity in graphene

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We describe the conditions under which the predicted (but not yet observed) zero-field inter-layer excitonic condensation in double layer graphene has a critical temperature high enough to allow detection. Both double monolayer and double bilayer graphene are considered in our theory. We present the calculation of the critical temperature for the inter-layer excitonic superfluid transition within the mean-field BCS theory for both optimistic and conservative approximations for the screening of the inter-layer Coulomb interaction, and discuss how disorder arising from charged impurities and corrugation in the lattice structure affect formation of the condensate via the charge inhomogeneity they induce. In the former case, we use a numerical Thomas-Fermi-Dirac theory to describe the local fluctuations in the electronic density in double layer graphene devices and estimate the effect these realistic fluctuations have on the formation of the condensate. We also estimate the effect of allowing dynamic contributions to the inter-layer screening. We then conduct similar calculations for double quadratic bilayer graphene, showing that the quadratic nature of the low-energy bands produces only a small qualitative change in the pairing strength. We compare the inter-layer interaction potential for these two systems and find that, since screening is stronger in quadratic bilayer graphene, it is probably not advantageous to search for the condensate in this system. We believe that disorder-free closely spaced double layers of monolayer graphene provide the most likely system for excitonic inter-layer superfluidity to manifest itself.

I. INTRODUCTION

The prediction of an inter-layer direct Coulomb interaction driven room-temperature excitonic condensate at zero magnetic field in double monolayer graphene (DMG) and double quadratic bilayer graphene (DQBG) has captured much attention both for the fundamental interest in the existence of a zero-field condensate and for the possible applications in devices, including ultra-fast switches and dispersionless field effect transistors. However, despite considerable experimental effort, the condensate has yet to be observed in Coulomb drag experiments in zero magnetic field. There are two possible reasons why this is so. First, the critical temperature \( T_c \) may simply be too small (i.e. much lower than the optimistic mean field theoretic predictions for ideal systems) and therefore the condensate is destroyed by simple thermal fluctuations. Second, it is possible that disorder in the form of electron–impurity scattering \( \tau_0 \) or from the presence of inhomogeneity in the charge distribution \( \rho \) is suppressing formation of the condensate.

To address the first of these possibilities, it is known that the size of the predicted excitonic gap in DMG depends very strongly on the choice of screening of the inter-layer Coulomb interaction. For DMG embedded in a dielectric medium such as hBN or SiO\(_2\), in the limit where the two layers have equal chemical potential but in opposite bands, and where \( k_F d = 0 \) (where \( k_F \) is the Fermi wave vector and \( d \) is the inter-layer separation), it is known that the prediction of room-temperature superfluidity is only valid for the unscreened Coulomb interaction approximation. Using static screening in the analysis (replacing the unscreened Coulomb interaction) exponentially suppresses the predicted excitonic gap to a regime where it is unmeasurable. A dynamic screening approximation gives a gap which is intermediate between these two and may exhibit a sharp increase if the effective interaction parameter \( \alpha = \frac{\epsilon^2}{(\hbar v_F)^2} \gtrsim 1.5 \), or equivalently \( \epsilon < 1.45 \) where \( \epsilon \) is the dielectric constant of the medium and \( v_F \) is the Dirac band velocity of monolayer graphene. The impact of vertex corrections has also been studied and is claimed to be weak, although they inevitably decrease the pairing strength. Inter-band processes have also been shown to slightly increase the critical temperature. In the context of quadratically dispersing bands, room temperature superfluidity was predicted for electron–hole systems with ordinary parabolic dispersion as early as 1976 by Lozovik and Yudson. For DQBG, it is claimed that there is a critical density of carriers below which screening is disrupted by the presence of the excitonic gap at the Fermi energy and even static screening can give \( T_c \) in the measurable range. This rationale (i.e. the self-consistent suppression of screening in the presence of the superfluidity gap itself) should apply equally well to monolayer graphene and semiconductor systems as well as for DQBG since there is nothing special about DQBG in this context. Thus, Ref. taken at face value, would indicate very high critical temperatures for excitonic condensation in double-layer semiconductor and DMG systems too, in direct conflict with experimental findings. Unfortunately, however, this approach relies on an erroneous diagrammatic expansion of the interaction and this claim is therefore somewhat dubious, as we show below. In particular, the correct theory for the screening function (i.e. the dielectric response) of a superconductor in the symmetry-broken phase is quite nontrivial and simply replacing the Green’s function in the polarizability by the correspond-
ing superconducting anomalous Green’s function, as done in Ref. [3], is manifestly incorrect since this violates the Ward identity, and one must include the infinite series of vertex corrections along with the self-energy corrections, a rather daunting task which has been taken on only rarely in the literature (see Refs. [17] and [18] for the details). How such a consistent approximation affects the excitonic condensation is unknown at this stage and remains an interesting open question. It is theoretically much more sensible at this stage to take a perturbative approach necessitating the use of the screened interaction in the non-symmetry-broken normal state rather than using an unjustified screening approximation (violating the Ward identity) in the superfluid phase.

The role of disorder has also been studied, and it was shown in Refs. [7-9] that intra-layer momentum scattering by short-range disorder typically does not reduce $T_c$ substantially. However, long-range disorder in the form of charge inhomogeneity does play a key role in this case the energy scale to which the disorder must be compared is the excitonic gap, which may be small depending on the choice of screening in the inter-layer interaction. The carrier density inhomogeneities induced by the presence of long-range disorder lead to differences in the chemical potential in the two layers that remove the perfect nesting of the Fermi surfaces and therefore strongly suppress $T_c$. This effect is analogous to the Clogston-Chandrasekhar limit of BCS superconductivity and the associated destruction of the pairing. It must be emphasized that although the usual momentum scattering by disorder does not affect the excitonic condensation by virtue of Anderson’s theorem, any density or chemical potential fluctuations between the two layers would act as a random magnetic field for the s-wave superconductor, strongly suppressing the superfluidity. Random charged impurities in the environment invariably lead to inter-layer chemical potential fluctuations because of their long-range Coulombic nature.

In this article, in Sec. II we give a comprehensive description of the conditions under which the condensate should be observable in DMG. To do this, we accomplish two main tasks. First, we describe how $T_c$ behaves in the presence of a finite imbalance in the chemical potentials of the two layers. We estimate $T_c$ for both the unscreened and statically screened inter-layer interactions as a function of the overall chemical potential ($\mu$) and asymmetry in the chemical potential in the two layers ($\delta\mu$), and provide estimates for the dynamic screening case. (Our reason for carrying out our calculations using both unscreened and statically screened Coulomb interactions is the fact that they respectively represent the most optimistic and the most pessimistic scenarios for the excitonic condensation to occur with the respective $T_c$ estimates for the two approximations differing by several orders of magnitude.) We find that any finite $\delta\mu$ has the effect of reducing $T_c$, and that when $\delta\mu$ is comparable to the size of the excitonic gap (\(\Delta\), $T_c$ becomes zero consistent with the Clogston-Chandrasekhar limit in metallic superconductors. The second task is to provide a comprehensive description of the nature of the charge inhomogeneity in DMG devices. We use Thomas-Fermi-Dirac theory (TFDT) to estimate the spatial fluctuation in $\delta\mu$ induced by randomly placed charged impurities. In this way, we gain a full understanding of the nature of the correlations in the disorder of the two layers, and obtain quantitatively accurate estimates of the spatial size and magnitude of the charge fluctuations for realistic experimental parameters. We then link the two calculations by assuming that the variance of the disorder-induced potential fluctuations represents the typical mismatch or imbalance of the Fermi energies in the two layers. We find that when charged impurities are located close to the DMG, the fluctuations in $\delta\mu$ have a length scale of \(\sim 10\) nm, and that $\delta\mu \sim \bar{\mu} > \Delta$ indicating that the excitonic condensate will not be able to form in this regime, as it is suppressed totally by the disorder induced inter-layer chemical potential fluctuations. However, when a clean spacer layer is used to separate the DMG from the SiO\(_2\) where the impurities reside, the fluctuations in $\delta\mu$ reduce by an order of magnitude for comparable impurity densities. In this regime, when the impurity density is low, we find that it is possible for the excitonic condensate to form for reasonable $T_c$ estimates as given by dynamic screening.

In Sec. III we present the condensate analysis of the DQBG system at the same level of approximation as that of the DMG. This is important since it provides a better qualitative and quantitative comparison to DMG than previous work on DQBG provided. We find that the alteration in the single particle band structure and chirality properties of the underlying layers in going from DMG to DQBG causes small quantitative changes in the stability of the predicted condensate in the unscreened interaction approximation. However, the effects of screening of the inter-layer interaction in DQBG are stronger than in DMG, meaning that for a realistic system, the predicted $T_c$ is smaller in DQBG. Our conclusion, that DMG is a better system to look for excitonic condensation than DQBG, is different from that in Ref. [3].

II. DOUBLE MONOLAYER GRAPHENE

A. $T_c$ for asymmetrically doped layers

We begin by describing the role of a finite chemical potential difference between the two layers in reducing $T_c$. A rigorous estimate of $T_c$ in the non-disordered case is already an intractable calculation since the inclusion of dynamic screening effects in the inter-layer interaction demands that the vertex corrections be included in the theory. In fact, purely technically, there is no Migdal’s theorem when considering superconductivity (or fermionic superfluidity) induced by electron-electron interactions, and thus vertex corrections must, in principle, be included in the theory even for the unscreened
interaction. As such, all mean-field BCS type theories of inter-layer excitonic superfluidity are somewhat suspect strictly from a theoretical viewpoint since vertex corrections are uncritically neglected in the BCS mean field theory. But in the presence of dynamic screening, the theory becomes particularly suspect if vertex corrections are left out since the interaction itself now is calculated in an approximation including infinite number of electron-hole loops. Therefore, since all analytically approachable calculations will either over-estimate (i.e. the unscreened case) or under-estimate (i.e. the statically screened case) the actual $T_c$, we shall show how the chemical potential imbalance will affect the formation of the excitonic condensate for two distinct model calculations. Using the unscreened inter-layer interaction (which will systematically over-estimate $T_c$), and the statically screened inter-layer interaction (which will under-estimate $T_c$), we show that when $\delta \mu > 0$ is less than the excitonic gap (which we label $\Delta$), $T_c$ is reduced but remains finite. When $\delta \mu \sim \Delta$, $T_c$ becomes zero. Thus, the disorder-induced chemical potential imbalance or asymmetry is a key parameter determining the existence or not of the excitonic condensation which needs to be taken into account in the experimental search for inter-layer superfluidity.

It is convenient to assume that the upper layer is doped with electrons, the lower layer with holes, and to perform a particle-hole transformation in the lower layer such that both layers are described by a chemical potential with positive sign. We then characterize the system by their average chemical potential $\bar{\mu} = (\mu_u + \mu_l)/2$ and difference $\delta \mu = \mu_u - \mu_l$. Note that in other works relating to the excitonic condensate in DMG, it was assumed that $\mu_u = \mu_l$ giving perfect nesting of the Fermi surfaces in the two layers.\(^{1,2,7,21,22}\) Within the mean-field theory and the BCS approximation, the temperature-dependent gap function is given by:^{10}

$$\Delta_{k}(T) = \sum_{k'} V(k-k') \frac{\Delta_{k'}(T) f(k,k') N(k',T)}{E_{k'}}.$$  \hspace{1cm} (1)

In this equation $V(q) = V(q,0)$ is the static limit of the random phase approximation (RPA) for the inter-layer potential, $f(k,k') = [1 + \cos(\theta_k - \theta_{k'})]/2$ comes from the chirality of electrons in monolayer graphene, $N(k',T) = n_\beta(k',T) - n_\alpha(k',T)$ is the finite temperature occupation factor of the excitonic bands labeled by $\alpha$ and $\beta$ which contain $\delta \mu$ and $E_{k'} = [(v_F k' - \mu)^2 + \Delta_{k'}^2]^{1/2}$ where $v_F$ is the monolayer graphene Fermi velocity. We identify the excitonic gap $\Delta$ as the peak value of this function, which, in the BCS limit, is found at $k = k_F$. We define $T_c$ as the lowest value of $T$ for which the gap function is zero for all $k$. We find this condition numerically, and it gives a value which is of the same order of the standard prediction $\Delta = 1.76T_c$ from the textbook constant gap approximation to mean field theory. Our value for $T_c$ is a little smaller due to the non-constant gap function and full momentum-dependent interaction which we retain in the calculation. (We note as an aside that in the presence of the Fermi surface mismatch in the two layers, i.e. a chemical potential or density imbalance between the layers, there can, in principle, be inhomogeneous FFLO type solutions for the ground state superfluidity in the system, but our general calculations allowing for the possibility of such inhomogeneous FFLO states fail to find any FFLO solutions for either the DMG or the DQBG systems, and we consistently find either purely homogeneous superfluid condensate or no condensate.)

The inter-layer screened interaction potential is calculated within the RPA, which is justified for double layer graphene because the fermion number $N = 8$ is large, as

$$V(q,\omega) = \frac{v_q e^{-q d}}{1 + 2v_q (\Pi_u + \Pi_l) + v_q^2 \Pi_u \Pi_l (1 - e^{-2q d})}.$$  \hspace{1cm} (2)

where $v_q = 2\pi e^2/(eq)$, $\Pi_u$ and $\Pi_l$ are the polarization functions for the upper and lower layers, respectively, and $q$ and $\omega$ are functions of $q$ and $\omega$. The polarization functions are given for both the static ($\omega = 0$) limit and dynamic approximations in Ref. 20. The unscreened and static screening cases represent the two limits for the size of the gap in a real system: the unscreened interaction tends to overestimate the pairing and hence gives a large $T_c$, while the static screening tends to overestimate the screening efficiency and therefore yields a small $T_c$. In this article, we consider both these cases making the reasonable assumption that the reality of the situation is somewhere in between.

In the optimistic case of the unscreened inter-layer
Coulomb interaction, close layer separation ($d = 1\text{nm}$), and a flat hBN substrate with $\epsilon = 3.9$ [see Fig. 1(a)] we find that $T_c$ can be of the order of 100K for realistic doping and moderate layer imbalance. As clarified in Fig. 1(c), $T_c$ is a monotonically increasing function of $\mu$, and larger $\delta \mu$ reduces $T_c$. Figure 1(d) shows that as the chemical potential asymmetry increases, $T_c$ decreases monotonically with a characteristic 'S' shape. Note that our theory neglects the possibility of direct tunneling between the layers, but this may become a significant factor and could even enhance the formation of the condensate\textsuperscript{15} as the inter-layer spacing becomes this small. Taking $d = 5\text{nm}$, which is thick enough to fully suppress inter-layer tunneling [see Fig. 1(b)], gives $T_c \approx 50\text{K}$. We have repeated these calculations using the statically screened inter-layer interaction finding that the gap is so small that $T_c$ is essentially zero even for $d = 1\text{nm}$ and $\epsilon = 1$. To demonstrate why, in Fig. 2(a) we show as a function of momentum $q$ the statically screened interaction normalized by the unscreened potential $V_0 = v_q e^{-q d}$. The potential is a universal function of $q/k_F$ for $d = 0$, but is weakly dependent on $k_F d$ for finite $d$. We show representative curves for $k_F d = 0$, $k_F d \ll 1$ and $k_F d > 1$ corresponding to the strongest interaction limit, the low density limit, and the high density limit respectively. We see that higher carrier density reduces the interaction strength since the increased density of states allows the screening to be more efficient. The static polarizability is constant up to $q = 2k_F$, but increases after that\textsuperscript{20} causing the noticeable flattening of the interaction potential as a function of $q$ in Fig. 2(a). The peak of the gap function is found at $k = k_F$ and the most relevant contribution to the integrand in Eq. (1) comes from $k = k' \approx k_F$ indicating that the important range of wave vectors is $|q| < k_F$. In this regime, the statically screened potential is an order of magnitude smaller than the unscreened one leading to an excitonic gap which is several orders of magnitude smaller than the unscreened case, consistent with previous analytical evaluations\textsuperscript{13}.

The inclusion of dynamic screening into the inter-layer interaction potential makes the full calculation of $T_c$ intractable. In addition, simply adding frequency dependence to the interaction, as in Eq. (2), is not a controlled approximation since the RPA neglects diagrams from the vertex correction which are of the same order as some of those included. It is also likely that these vertex corrections will substantially reduce the inter-layer interaction potential although some authors claim otherwise\textsuperscript{12}. However, since this form of the interaction has been used by other authors, we analyze the potential function Eq. (2) itself to gain some intuitive insight into the effect of dynamic screening. In Fig. 2(b) we show the dynamically screened interaction potential normalized by $V_0$ as a function of wave vector for various frequencies and $k_F d = 0$. In this limit, the interaction is a universal function of $q/k_F$. The dynamic screening is a strong function of the frequency $\omega$, however some instructive patterns can be identified. For $q/k_F < \omega/\bar{\mu}$, the polarization functions are negative\textsuperscript{20} and therefore it is possible for the finite frequency potential to have a divergence corresponding to the plasmon wave vector $k_p$. The potential also is negative\textsuperscript{21} for $k_p/k_F < q/k_F < \omega/\bar{\mu}$ indicating an overall repulsive interaction which could reduce the gap size. In contrast, we note that for $q < k_p$, the interaction is enhanced over the unscreened case. This is the well-known anti-screening effect of dynamic screening, which should to some extent compensate for the sign change of the interaction in some regime of the phase space. For $q/k_F > \omega/\bar{\mu}$, the potential is very similar to the statically screened case. The most relevant frequency range is $\omega \approx \Delta$, which is in general rather small compared to $\bar{\mu}$ and hence the range of wave vectors where the potential deviates substantially from the statically screened case is small, indicating that $T_c$ with dynamic screening will be closer to that predicted by the static screening calculation than the unscreened one. This analysis also shows that a system with a large bare gap will be more robust against dynamic screening effects since the relevant frequency will be higher, implying that within the range of $q$ that contributes strongly to the integrand in Eq. (1) we have $V(q, \omega)/V_0 > 1$. It may be appropriate to emphasize here again that one cannot account for the superfluid gap in the screening simply by adding the self-energy to the Green’s function in the polarizability bubble, although it is fairly simple to do so, since this would violate the Ward identity, and thus be a manifestly incorrect approximation\textsuperscript{22}.

Calculations for the unscreened interaction for suspended DMG with $\epsilon = 1$ and $d = 1\text{nm}$ show that the excitonic gap is large with respect to $\bar{\mu}$ and therefore fluctuations of the order of the chemical potential will not reduce $T_c$ to zero. When $d = 5\text{nm}$ we find that $T_c$ drops to room temperature or a little below. Suspended graphene is also known to form ripples with size fluctuations of the order of 1nm in height\textsuperscript{22} which may make the precise control of the inter-layer spacing difficult for these proposed devices, and which may introduce charge inhomogeneity related to the strain field induced by the corrugations. We shall discuss the effects of ripples in the
next section, after we have described the role of charge inhomogeneity in DMG.

B. Charge inhomogeneity in DMG

In any experimental sample, some degree of extrinsic disorder-induced charge inhomogeneity will exist, as has been demonstrated by many surface measurements of monolayer graphene.\textsuperscript{22,25} In a double layer device, the inhomogeneities in the charge landscapes will not be identical in both layers, and therefore there will be spatial variation in the asymmetry of the chemical potentials. In this situation, the local difference in chemical potential has two contributions. There is a nominally homogeneous part which is induced by gating and is in principle controllable. This contribution was the subject of the previous section and we ignore it here. Then there is a contribution from charged impurities and other disorder which is inhomogeneous and not controllable. A full analytical description of the inhomogenous system is clearly intractable so we employ an accurate numerical method to compute the charge density of the system when charged impurities explicitly break translational symmetry. From this charge landscape, we can assign the local chemical potential $\mu_i(\mathbf{r})$ and $\mu_l(\mathbf{r})$ in each layer, and characterize the spatial fluctuations by their root-mean square (rms) value, which is a measure of the typical fluctuation. Using this measure of the disorder in the charge landscape, we can discuss the stability of the condensate against the density and chemical potential inhomogeneity induced by the charged impurities.

To calculate the charge landscape in each layer taking into account the presence of long-range disorder due to the charged impurities and nonlinear screening effects we use the TFDT.\textsuperscript{26} The TFDT is a generalization to Dirac materials of the Thomas-Fermi theory (TFT).\textsuperscript{26} TFDT and density-functional-theory (DFT)\textsuperscript{27–29} are both non-perturbative functional theories and therefore have the great advantage of being able to take into account nonlinear screening effects that are dominant in systems like graphene at low doping. TFT and TFDT differ from DFT in that in these theories the kinetic energy term is also treated via a functional. As a consequence TFDT is computationally much more efficient than DFT and can be used to study large systems in the presence of long-range disorder where DFT is completely impractical. In particular by using the TFDT we are able to obtain disorder-averaged results which would be impossible with the strict DFT approach because of the heavy numerical cost. The simplified treatment of the kinetic energy term limits the validity of TFDT to regimes in which $|\nabla n/n| < k_F$. We have verified that this condition is reasonably well satisfied in single layer graphene.\textsuperscript{23–24}\textsuperscript{31} Our results show that in graphene double layers, due to the increased screening of the disorder potential caused by the presence of an additional graphene layer, the correlation length of the disorder-induced inhomogeneities is larger than in isolated single layer graphene and therefore that the condition $|\nabla n/n| < k_F$ is always well satisfied in these double layer graphene heterostructures.

The carrier density ground state is obtained by minimizing the TFDT energy functional

$$E[n_u, n_l] = E_u[n_u(\mathbf{r})] + E_l[n_l(\mathbf{r})] + \frac{e^2}{2\kappa} \int d^2r \int d^2r' \frac{n_u(\mathbf{r})n_l(\mathbf{r}')}{(|\mathbf{r} - \mathbf{r}'|^2 + d^2)^{1/2}}$$

where $E_l[n_l(\mathbf{r})]$ is the energy functional of the density profile for the $i$-th graphene layer (as given in Ref. 19) and the last term is the inter-layer Coulomb interaction. Each layer functional $E_i[n_i(\mathbf{r})]$ contains a term due to the disorder potential $V_D$ created by the charged impurities. We also include intra-layer exchange interactions. We assume that charged impurities located close
to the surface of the SiO$_2$ constitute the dominant source of disorder and we therefore model the charged impurity distribution as an affective two-dimensional distribution $C(r)$ placed at a distance $d_B$ below the lower graphene layer. Note that this is the most generous estimate for the charged impurities that we can take, since we are neglecting any disorder at the other interfaces which may be induced by the successive fabrication steps required to make these devices. Denoting disorder-averaged quantities by angle brackets, without loss of generality, we assume $\langle C(r) \rangle = 0$. We also assume the charged impurities to be uncorrelated so that $\langle C(r)C(r') \rangle = n_{\text{imp}} \delta(r - r')$, where $n_{\text{imp}}$ is the charged impurity density. $V_D$ in each layer is the Coulomb potential created by the random distribution $C(r)$. The ground state density distributions $n_u(r)$ and $n_l(r)$ are obtained by minimizing $E[n_u, n_l]$ numerically enforcing the self-consistency of the distribution in the two layers due to the inter-layer interaction. Then, the local difference in chemical potential between the two layers $\delta \mu_{\text{loc}} = \mu_u(r) - \mu_l(r)$ can be extracted for each point in the system and by performing the minimization for many ($\sim 600$) disorder realizations and we obtain statistics for the distribution function of $\delta \mu_{\text{loc}}$.

In Fig. 3 we show the spatial distribution of $\delta \mu_{\text{loc}}$ for three different impurity densities and in two experimentally relevant geometries. In the left column, we show data where the lower graphene layer is placed directly onto an SiO$_2$ substrate and the inter-layer separation is $d = 1$nm. In this case, the charged impurities at the oxide interface are effectively approximated by a two-dimensional distribution 1nm below the lower graphene layer, so we take $d_B = 1$nm. This corresponds to the system used in the Austin experiments. We set $\bar{\mu} = 50$meV corresponding to an easily achievable carrier density regime. The right column corresponds to a system where the lower graphene layer is separated from the SiO$_2$ substrate by a 20nm layer of hBN, like that used in the Manchester experiments. Hence, we take $d_B = 20$nm, and we also set $\bar{\mu} = 200$meV corresponding to the high carrier density regime where we expect the screening of the external impurities to be the most efficient, resulting in the lowest amount of charge inhomogeneity. In both situations we assume that the gate-induced (homogeneous, controllable) part of the layer asymmetry to be zero. Therefore we assume that any layer imbalance is completely defined by the charged impurities. The rows of Fig. 3 show the data for, from top to bottom, $n_{\text{imp}} = 10^{11}$cm$^{-2}$, $n_{\text{imp}} = 10^{10}$cm$^{-2}$, and $n_{\text{imp}} = 10^9$cm$^{-2}$. (We note that the higher value of $n_{\text{imp}}$ is more typical, and $n_{\text{imp}} = 10^9$cm$^{-2}$ is unlikely to be achieved in laboratory graphene samples on any substrate. Typically, one can get an estimate of $n_{\text{imp}}$ in a particular sample by looking at the carrier density regime over which the graphene minimum conductivity ‘plateau’ exists around the Dirac point.) All six plots share some similar qualitative features. In particular, they all show regions where $\delta \mu$ is positive and regions where it is negative with narrow strips in between where $\delta \mu$ is small. The lengthscale of the fluctuations is not affected by the impurity density, but the magnitude of the fluctuations is. By comparing the data in the two columns we see that the distance of the impurities from the DMG makes a substantial difference in the length scale of the fluctuations in $\delta \mu_{\text{loc}}$, and in reducing the magnitude of the fluctuations. The density of impurities also has a strong effect on the magnitude of the fluctuations in $\delta \mu_{\text{loc}}$ with the fluctuations reducing by approximately a factor of three with each order of magnitude decrease in $n_{\text{imp}}$. In the most dirty case (it should be noted that graphene on SiO$_2$ can have an impurity density of up to $5 \times 10^{12}$cm$^{-2}$ as measured by transport measurements), shown in Fig. 3(a) then the fluctuations in $\delta \mu_{\text{loc}}$ may be of the order of $\bar{\mu}$, indicating that the condensate has no opportunity to form in this case. For the cleanest situation shown in Fig. 3(f), the potential imbalance is on the scale of 1meV and there is a significant chance that excitons with a gap of the size predicted by dynamic screening calculations will persist in spite of the disorder if an impurity density as low as $10^9$cm$^{-2}$ can be achieved in double layer graphene samples.

We take many disorder realizations ($\sim 600$) for each impurity density and collect ensemble-averaged statistics for the distribution of $\delta \mu_{\text{loc}}$. We characterize this distribution by its root-mean-square value, which we label $\delta \mu_{\text{rms}}$. In Fig. 4 we plot $\delta \mu_{\text{rms}}$ for the two experimen-
tional geometries discussed above and for different inter-layer spacing $d$. This is shown in Figs. 1(a) and 1(b) for $d = 1$ nm and $d = 5$ nm, respectively for three impurity densities covering three orders of magnitude. The fluctuations are strongest at low carrier density, where the screening of the impurity potential is weakest, and it decreases monotonically with increasing $\bar{\mu}$. The trend suggested by the spatial plots is confirmed here, that decreasing the impurity density by a factor of ten generates approximately a factor of three reduction in the fluctuations. If the impurities are moved away from the DMG by a spacer layer as in Figs. 1(c) and 1(d) we find that the fluctuations in $\delta \mu$ are reduced to the order of 1 meV. This degree of fluctuation may be small enough to allow the condensate to be detected at a reasonable temperature scale.

We briefly discuss the role of corrugations and ripples in the structure of the graphene lattice and describe their effect on the excitonic condensate. The existence of these ripples in suspended samples and those placed on substrates of different kinds have been demonstrated. In the case of suspended monolayer graphene, ripples of height of several angstroms have been observed indicating that a suspended double layer structure with small inter-layer spacing may be difficult to control since ripple corrugations will then be of the same size as the layer separation. It has been theoretically predicted that the strain field associated with ripples can give rise to fluctuations in the local density of the order of $10^{12}$ cm$^{-2}$. If this is correct then it shows that lattice corrugations may be a significant barrier to the existence of the condensate, since this will give $\delta \mu \approx 40$ meV which would destroy the condensate even for unscreened Coulomb interactions.

III. DOUBLE BILAYER GRAPHENE

We now consider the analogous situation for double bilayer graphene (DQBG). In this case, two AB-stacked bilayer graphene sheets replace the monolayers discussed previously. We employ the same approach as we did for DMG in that we analyze the critical temperature of the excitonic superfluid for the unscreened and statically screened inter-layer interactions, and then examine the modification of the inter-layer potential with the inclusion of dynamic screening effects. We model the two bilayer graphene sheets as having a gapless, quadratic low energy band structure $E_{\nu k} = \nu \hbar^2 k^2/(2m^*)$ where $m^*$ is the effective mass which we assume to be the same in both layers, and $\nu = \pm 1$ denotes the band. Note that this approximation is only valid at relatively low densities, $n < 3 \times 10^{12}$ cm$^{-2}$ which is approximately equivalent to $k_F = 0.3$ nm$^{-1}$ and $\mu \approx 70$ meV. The gap equation in Eq. (1) and inter-layer interaction in Eq. (2) are still valid in the double bilayer case, except for a redefined angular factor $f(k,k') = \cos^2(\theta_k - \theta_{k'})/4$, exciton energy $E_{k'} = [(\hbar^2 k'^2/(2m^*) - \bar{\mu})^2 + \Delta^2_{k'}]^{1/2}$, and the polarization screening function of the quadratic bilayer obviously needs to be taken into account.

We first show results for the unscreened interaction, using $V(q) = 2\pi e^2 c^{-q} / (q \kappa)$. In Fig. 2(a) we show $T_c$ as a function of $\bar{\mu}$ and find that it is of the same order as for the equivalent case in DMG, i.e. $T_c \sim 100K$ for realistic inter-layer separation [c.f. Fig. 1(a)]. However, the non-monotonicity of $T_c$ as a function of overall density is unlike the monolayer case indicating that the inter-layer separation and density of electrons have a more complex relationship than in the linear spectrum. This is due to the effective interaction parameter $r_s$ being constant for monolayer graphene, but decreasing as $1/\sqrt{n}$ as a function of density for bilayer graphene. Therefore, as density increases, the reduced strength of the interactions in DQBG manifests as a smaller excitonic gap, thus reducing $T_c$. Figure 2(b) shows $T_c$ as a function of $\delta \mu$ for $\bar{\mu} = 30$ meV. This shows qualitatively similar behavior as for DMG, indicating that within our BCS mean field theory, the details of the underlying band structure do not qualitatively affect the response of the excitonic superfluid to asymmetrical layer doping.

As shown in Ref. 12, the static screening for quadratic bilayer graphene is somewhat stronger than for monolayer graphene, not just because the dimensionless polarizability is larger, but because the density of states in the prefactor is also larger at moderate doping. This indicates that the inter-layer interaction should be weaker in DQBG compared with DMG for comparable parameters. We demonstrate this in Fig. 5(c) for the statically screened interaction. In contrast to the DMG case, the inter-layer interaction $V(q,0)$ given in Eq. (2) is not a universal function of $q/k_F$ for DQBG in the $k_Fd = 0$ limit. Increasing the electronic density decreases the efficiency of the screening and allows the inter-layer interaction to be stronger. This is a substantial qualitative difference between the screening in monolayer and bilayer graphene which is understandable since quadratic bilayer has a constant density of states in contrast to the linear-in-energy density of states of monolayer graphene. The kink at $q = 2k_F$ corresponds to the $2k_F$ anomaly for $\Pi(q)$. However, increasing $d$ allows the screening to be more efficient, reducing $V(q,0)$ with respect to $V_q$, as in DMG.

For dynamic screening, we use previous results for the finite frequency polarizability to determine the interaction potential. Figure 5(d) shows the dynamically screened interaction in the high density limit, where the interaction is the strongest. The high-$q$ limit is the same as the static screening case, and we find that the dynamic screening reaches this limit even faster than the DMG. Therefore, the interaction strength is weaker in DQBG and $T_c$ is suppressed, making the observation of the excitonic superfluid less likely in this system compared to DMG.

We now briefly comment on the role of inhomogeneity in DQBG. Since it is known that the response of the charge distribution to charged impurities in bilayer graphene is qualitatively similar to that in mono-
layer graphene, it is very likely that the presence of charged impurities in the environment of the DQBG will have a similar detrimental effect on the stability of the condensate as for DMG. However, it is also known that bilayer graphene is, in general, somewhat more robust against ripples and corrugations than monolayer graphene, indicating that charge inhomogeneity generated by this form of disorder may be slightly less important.

Recently, the authors of Ref. 3 claimed that room-temperature superfluidity is possible in DQBG by using a screening model which includes the existence of superfluidity with the associated gap at the Fermi energy. In this theory, the gap at the Fermi energy reduces static screening to a minimal level, enabling the condensate to be stable at temperatures of the order of 100K. In fact, there is no reason why this argument cannot be applied to DMG or regular semiconductor bilayer systems since the superfluid gap according to such arguments would always suppress screening at the Fermi level. However, no compelling evidence of excitonic condensation has been seen at zero magnetic field in DMG or semiconductor double-layer systems calling into question this approach based on the proposed theoretical suppression of screening by the superfluid gap itself. As we discussed earlier in this paper and as described in great details in Refs. 17 and 18, the theoretical formulation of the dielectric response of a superconductor in the gapped symmetry-broken phase is a formidable task, which cannot be simulated simply by incorporating the self-energy in the polarization bubble diagram since this would manifestly violate the Ward identity. It is therefore theoretically more meaningful to use a perturbative approach where the screened interaction is calculated in the normal state (as done in the current paper) and then the BCS mean field theory is carried out on this normal state interaction function. In the current manuscript, we have demonstrated that DMG and DQBG have the same level of approximation for the inter-layer screening. We do not believe that DQBG offers any advantage over DMG in the realization of the excitonic superfluid phase. Additionally, we have shown that screening is in fact stronger in DQBG than in DMG at a given level of approximation, indicating that DMG is a more promising system for the observation of the condensate. 

From a technical point of view, the authors of Ref. 3 use a diagrammatic expansion of the inter-layer interaction which is not well defined. Specifically, it has been shown that to self-consistently include the condensate in the inter-layer screening, a full dynamical calculation of the vertex correction must be undertaken, and that the effect of the vertex corrections is to reduce the interaction strength considerably. In contrast, the analysis we present is properly defined in the sense that all diagrams of the same order are included in the perturbation theory. Our work shows that the key to the laboratory realization of graphene double layer excitonic condensation is sample purity — in particular, all inhomogeneous inter-layer density fluctuations must be minimized by having ultra-pure graphene layers free of ripples and random charged impurities for there to be any hope of discovering the inter-layer excitonic superfluid phase.

IV. CONCLUSION

In summary, we have presented a comprehensive analysis of $T_c$ for the excitonic condensate in DMG, and described the conditions under which the condensate may be observable in experiments. We find that the existence of charge inhomogeneity and ripples is likely to be the limiting factor for the stability of the condensate, but that the cleanest samples at low temperature should allow for the detection of the condensate. If the graphene layers are suspended, particular care must be taken to ensure minimal rippling since even in the absence of charged impurities, this may be a significant source of charge inhomogeneity. We also investigated similar physics in DQBG, showing that the quadratic nature of the low energy bands has a small, quantitative effect on the $T_c$ of the condensate in the absence of inter-layer screening. However, since screening is stronger for bilayer graphene than for monolayer graphene, we find that $T_c$ for a realistic system will be smaller in DQBG than in DMG for similar system parameters. Calculating the gap using the static screening RPA approximation, we demonstrated that the gap is vanishingly small in the double bilayer case, as in the double monolayer case. Our most
important conclusion is that the only hope for achieving excitonic condensation in graphene is to use ultra-pure graphene with very low disorder and ripple induced charge inhomogeneity. Even then, if the operative inter-layer pairing interaction turns out to be the statically screened Coulomb interaction, there is very little hope for the observation of the inter-layer superfluid state at any reasonable finite temperatures.

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