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Disorder-tuned selection of order in bilayer graphene

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The nature of the interaction-driven spontaneously broken-symmetry state in charge-neutral bilayer graphene (BLG) has attracted a lot of interest. Theoretical studies predict various ordered states as the candidates for the ground state of BLG in the absence of external fields. Several experiments have been performed by different groups to identify the nature of the collective ground state in BLG. However, so far, there is no consensus: some experiments show evidence that suggests the establishment of a nematic gapless state, while others present results that are more consistent with the establishment of a fully gapped state. Moreover, even among the experiments that appear to see a bulk gap, some of the samples are found to be conducting (suggesting the existence of gapless edge states), while others are insulating. Here we explore the hypothesis that disorder might explain the discrepancy between experiments. We find that the pair-breaking effect due to nonmagnetic short-range disorder varies among the candidate ground states, giving rise to different amounts of suppression of their mean-field transition temperatures. Our results indicate that BLG can undergo a transition between different ordered states as a function of the disorder strength, providing a possible scenario to resolve the discrepancy between experimental observations.

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I. INTRODUCTION

AB-stacked bilayer graphene (BLG) [1–4] is formed by two graphene [5] layers rotated by 60° with respect to each other. Its low-energy band structure is characterized by parabolic conduction and valence bands that touch at the corners, the K and K′ points, of the Brillouin zone. A number of theoretical works have predicted various spontaneously-broken-symmetry states as the candidates for the ground state of BLG near the charge-neutrality point (CNP) in the absence of external fields [6–18] and related systems [19]. The multiple degrees of freedom in BLG—layer, spin, and valley—give rise to the diversity of the candidate orders. In general, the proposed ordered states can be classified in two groups: (i) gapped states characterized by the opening of a full gap in the quasiparticle spectrum, and (ii) nematic states in which the quadratic band crossing points at which the conduction and valence bands touch are split into two Dirac points, leaving the quasiparticle spectrum gapless. These two groups have a different structure with respect to the layer index: gapped states are layer-polarized while nematic states are not [15]. Depending on the valley and spin structure, different collective states can be identified in each general group. Gapped states with different spin-valley structures include the quantum valley Hall (QVH), the quantum anomalous Hall (QAH), and the quantum spin Hall (QSH) state, as well as a layer antiferromagnet (LAF) state. Within mean-field theory, in the clean limit, the states in each group have the same transition temperature: $T_{c,0}^G$ for the gapped states, and $T_{c,0}^N$ for the nematic states.

Several experimental groups have made efforts to ascertain the nature of the ground state by using high-quality suspended BLG [20–27]. They all find evidence of spontaneous symmetry breaking at low temperatures. However, they reach different conclusions on the identity of the ordered state: First, some experiments show evidence that supports the establishment of a nematic state [22], while others either present results that are more consistent with the establishment of a gapped state [23–27] or are consistent with both types of states [20,21]. Second, among the experiments supporting the establishment of a gapped state, some indicate that the gapped state comes with conducting edge states [20,21,23,27] and others indicate that the state is fully insulating [23–27], e.g., the LAF state. One explanation that has been proposed for this multitude of conflicting experimental results is that BLG is highly multicritical [28], and that different experimental samples fall in the basin of attraction of different correlated fixed points.

One important and unavoidable factor present in all materials that has the potential to strongly affect the formation and nature of a broken symmetry state is disorder due, for instance, to charge impurities, adatoms, vacancies, and ripples. For example, it is well known that the presence of magnetic impurities in BCS superconductors can strongly decrease the transition temperature ($T_c$) [29,30]. The pair-breaking effect of magnetic impurities in BCS superconductors can be attributed to the different scattering off the impurities of the time-reversed fermionic states forming the Cooper pairs. Another example is the pair-breaking effect of normal impurities on exciton condensates [31,32]. Since the broken-symmetry states in BLG involve particle-hole pairing with different layer-spin-valley structures, we expect that different pairing structures could be affected differently by disorder.

In this work, we study the effect of disorder on the broken-symmetry states of BLG near the CNP in the absence of external fields. We consider only nonmagnetic disorder and do not take into account spin-flip scattering. Within mean-field theory, in the clean limit, the transition temperature of the gapped phase is higher than that of the nematic phase. However, we find that this scenario can be modified when the presence of disorder is taken into account. Considering nonmagnetic short-range disorder, we find that, in the presence of disorder that causes intravalley scattering only, the transition
functions of the intravalley disorder strength by solving Eq. (8) for the case of uncorrelated disorder in which $\delta_N = b_0/2$. In this case that the clean-limit transition temperatures of the two phases have the relation $T_c^N/T_c^G = 0.8$. (b) Phase diagram obtained by calculating the critical disorder strength for various ratios of $T_c^N/T_c^G$ for the case of uncorrelated disorder in which $\delta_N = b_0/2$.

The influence of disorder is taken into account by using the self-consistent Born approximation. After averaging over disorder realizations, the effect of disorder is captured by the self-energy matrix $\Sigma$ that renormalizes the quasiparticle propagator and the pairing vertex of the condensate.

### A. Intravalley disorder scattering

We first consider the case in which disorder-induced valley-flip scattering processes are negligible, i.e., $\bar{W} = 0$. In this case, our discussion can be simplified to the $2 \times 2$ layer space since intravalley scattering does not lift the degeneracy between ground states that differ in valley structure. The renormalized Green’s function $\hat{G}$ is given by

$$\hat{G}(k, i\omega_n) = [i\omega_n \delta_0 - \hat{h}(k) - \hat{\Delta} - \hat{\Sigma}(k, i\omega_n)]^{-1}, \quad (2)$$

where $\omega_n = (2n + 1)\pi T$ are the Matsubara frequencies, $T$ is the temperature, and

$$\Sigma_{\sigma\sigma'}(k, i\omega_n) = n_{\sigma} \int \frac{d^2p}{(2\pi)^2} U_{\sigma, k-p} \hat{G}_{\sigma\sigma'}(p, i\omega_n) U_{\sigma', p-k} \quad (3)$$

is the disorder-averaged self-energy. Here $n_{\sigma}$ is the density of the randomly distributed intravalley scattering centers. It is reasonable to assume $n_{\sigma}$ to be the same in the two layers.

For the gapped states, the self-consistency equation for the order parameter takes the form

$$\Delta_G = -\frac{1}{2} \Gamma_{kz} \sum_n \int \frac{d^2k}{(2\pi)^2} \text{Tr}[\hat{\rho} \hat{G}(k, i\omega_n)], \quad (4)$$

where $\Gamma_{kz}$ is the effective coupling and $\text{Tr}[\ldots]$ is the trace of the argument. The disorder renormalized Green’s function can be written as

$$\hat{G}_G(k, i\omega_n) = \left[ i\omega_n - \Delta_G - \epsilon_k e^{i2\theta} \begin{bmatrix} -\mu & \epsilon_k e^{-i2\theta} \\ \epsilon_k e^{i2\theta} & -\mu \end{bmatrix} \right]^{-1}, \quad (5)$$

where

$$\bar{\omega}_n = \omega_n + n_{\sigma} \int \frac{d^2p}{(2\pi)^2} |U_{\sigma, k-p}|^2 \frac{\tilde{\omega}_\sigma}{\tilde{\omega}_\sigma^2 + \epsilon_p^2 + \Delta_G^2} \quad (6)$$

In the above expressions we have assumed that the disorder strength is the same in the two layers, i.e., $|U_{k-p}| \equiv |U_{1-k-p}| = |U_{2-k-p}|$. In the case of a short-range disorder potential,
$U_{\sigma,k-p} = U$, we obtain
\[
\tilde{\omega}_n = \omega_n + \frac{1}{2} \left( \frac{1}{\tau_2} + \frac{1}{\tau_1} \right) \tilde{\omega}_n, \\
\tilde{\Delta}_G = \Delta_G - \frac{1}{2} \left( \frac{1}{\tau_2} - \frac{1}{\tau_1} \right) \tilde{\Delta}_G,
\]
(7)
where $\frac{1}{\tau_1}$ and $\frac{1}{\tau_2}$ are the collision rates resulting from the disorder potential. In this case, $\frac{1}{\tau_1} = n_U U^2 m^* 2\pi^2$ and $\frac{1}{\tau_2} = 0$. Note that the opposite sign in front of $\frac{1}{\tau_2}$ in the equations for $\tilde{\omega}_n$ and $\tilde{\Delta}_G$ gives rise to the pair-breaking effect of disorder on the condensate. On the other hand, the term proportional to $\frac{1}{\tau_1}$ in Eq. (7) has the same sign in the equations for $\tilde{\omega}_n$ and $\tilde{\Delta}_G$ and, consequently, $\frac{1}{\tau_1}$ does not affect the transition temperature. We can therefore see that, for the gapped state, the effect of intrav valley disorder is analogous to the effect of magnetic impurities on BCS superconductors [29], which is purely pair breaking. From Eqs. (4) and (7) the mean-field critical temperature $T_c$ in the presence of disorder is given by a universal function in terms of the pair-breaking parameter $\delta = 1/\tau_2$ [29],
\[
\ln \left[ \frac{T_{c,0}}{T_c} \right] = \psi \left( \frac{1}{2} + \frac{\delta}{2\pi T_c} \right) - \psi \left( \frac{1}{2} \right),
\]
(8)
where $\psi(z)$ is the digamma function, and $T_{c,0}$ is the transition temperature in the clean limit. For the gapped phase, $T_{c,0} = T_{c,0}^G$ is given by
\[
k_B T_{c,0}^G = \frac{2}{\pi} \gamma E_c \exp \left[ -\frac{4\pi h^2}{\Gamma s m^*} \right],
\]
(9)
where $\gamma \approx 1.78$ is Euler’s constant, and $E_c$ is a cutoff for the energy range of the interaction. The value of the pair-breaking parameter $\delta$ is $\delta_G = \frac{1}{\tau_1} = n_U U^2 m^* 2\pi^2$ for the gapped states. When $\delta_G/2\pi T_c \ll 1$, the transition temperature is linearly suppressed: $T_{c,0}^G = T_{c,0}^G - \frac{\delta_G}{2}$. The critical disorder strength above which the gapped phase is completely suppressed, is given by $\delta_G^C = \frac{\pi}{2} (2\pi T_c)^2 \approx 0.887 T_{c,0}^G$. Assuming that the dominant source of disorder is charge impurities [4] and using the condition $\delta_G^C = 0.887 T_{c,0}^G$, we can provide a quantitative estimate of the critical value of the impurity density $n_{imp,c}$ above which $T_c \rightarrow 0$. Taking into account screening effects, the effective, screened, disorder potential $V_{D,sc}$ due to the charge impurities is short range with strength $U(q) = V_{D,sc}(q) = 2\pi e^2/(k q \epsilon(q))$, where $k$ is the dielectric constant and $\epsilon(q)$ is the dielectric function. For $q < 2k_F$ we have [4] $U = V_{D,sc}(q < 2k_F) = 2\pi e^2/(g_s g_v m^*)$ where $g_s = g_v = 2$ are the spin and valley degeneracy, respectively. We then find (set $k_B \equiv 1$ here) the critical impurity density
\[
n_{imp,c}^G = \frac{4}{\sqrt{\pi}} \frac{m^*}{h^2} T_{c,0}^G = (3 \times 10^{10}) \text{ cm}^{-2}. \text{meV}
\]
(10)
Experimentally, for the gapped phase $T_{c,0}^G$ appears to be on the order of 1 meV [25]. Equation (10) then allows us to predict that, in order to have the establishment of the gapped phase, the impurity density has to be lower than $\sim 3 \times 10^{10}$ cm$^{-2}$. This estimate is consistent with current experiments; see in particular Refs. [20,21]. In addition, Eq. (10) allows us to obtain $T_{c,0}^G$, a quantity that is very difficult to estimate accurately, by knowing the value of $n_{imp,c}^G$.

For the nematic states, the self-consistent equation for the order parameter takes the form
\[
\Delta_N = -\frac{1}{2} \Gamma D T \sum_n \int \frac{d^2k}{(2\pi)^2} \text{Tr} [\hat{\sigma}_2 \hat{G}(k, i\omega_n)],
\]
(11)
where $\Gamma_D$ is the effective interlayer coupling. The renormalized Green’s function after averaging over disorder can be written as
\[
\hat{G}_N(k, i\omega_n) = \left[ \frac{-i\tilde{\omega}_n - \Delta_N}{-\epsilon_k e^{i\omega_n} - \Delta_N} \right]^{-1},
\]
(12)
where
\[
\tilde{\omega}_n = \omega_n + n_U \int \frac{d^2p}{(2\pi)^2} |U_{k-p}|^2
\]
\[
\times \frac{\tilde{\omega}_n}{\tilde{\omega}_n^2 + \epsilon_p^2 + \Delta_N^2 + 2\epsilon_p \Delta_N \cos(2\theta_p)},
\]
\[
\tilde{\Delta}_N = \Delta_N - n_U \int \frac{d^2p}{(2\pi)^2} U_{k,p} U^*_{k+p}
\]
\[
\times \frac{\epsilon_p e^{-i\omega_n}}{\epsilon_p^2 + \Delta_N^2 + 2\epsilon_p \Delta_N \cos(2\theta_p)}.\]
\]
Here again we assumed $|U_1| = |U_2|$. In order to discuss the influence of disorder on $T_c$, we evaluate Eq. (13) in the limit $T \rightarrow T_c$, where the order parameter becomes vanishingly small, $\Delta_N \rightarrow 0$. Assuming short-range disorder, $U_{\sigma,k-p} = U_c$, to leading order in $\Delta_N$ we obtain (for $\tilde{\omega}_n > 0$)
\[
\tilde{\omega}_n = \omega_n + \frac{1}{2} \left( \frac{1}{\tau_2} + \frac{1}{\tau_1} \right) \tilde{\omega}_n, \\
\tilde{\Delta}_N = \Delta_N - \frac{1}{2} \left( \frac{1}{\tau_2} - \frac{1}{\tau_1} \right) \tilde{\Delta}_N.
\]
(14)
Linearizing Eq. (11) near $T_c$, we again find that the transition temperature satisfies Eq. (8), with the pair-breaking parameter $\delta_N = \frac{1}{\tau_1}$. In the limit $\delta_N/2\pi T_c \ll 1$, the transition temperature is linearly suppressed: $T_{c,0}^N = T_{c,0}^G - \frac{\delta_N}{2}$. The critical disorder strength above which the nematic phase is completely destroyed is given by $\Delta_N^C \approx 0.887 T_{c,0}^G$. Notice that both the clean-limit transition temperature and the value of the pair-breaking parameter are different from those obtained for the gapped phase. For the nematic phase, the mean-field transition temperature in the clean limit is given by
\[
k_B T_{c,0}^N = \frac{2}{\pi} \gamma E_c \exp \left[ -\frac{8\pi h^2}{\Gamma s m^*} \right].
\]
(15)
Notice that, assuming $\Gamma_D \approx \Gamma_S$, Eqs. (15) and (9) imply $T_{c,0}^N < T_{c,0}^G$. Equation (13) shows that the renormalized quantity $\tilde{\Delta}_N$ depends on the correlation property between the disorder potentials in the two layers: (i) When the disorder potentials in the two layers are perfectly correlated, $U_1 = U_2 \equiv U$, we have $\frac{1}{\tau_1} = n_U U^2 m^* 2\pi^2$, $\frac{1}{\tau_2} = n_U U^2 m^* 2\pi^2$, so that $\Delta_N = n_U U^2 m^* 2\pi^2$. In this case the relation between the pair-breaking parameter
values in the two phases is $\delta_N = \frac{3}{2} \delta_G$. (ii) When the disorder potentials of the two layers are uncorrelated, $\Sigma_{12} = \Sigma_{21} = 0$, then $\frac{1}{\tau_i} - \frac{1}{\tau_i} = 0$. In the limit $T \to T_c$, $\frac{1}{\tau_i} = \frac{1}{\tau_i} = n_U U^2 \frac{m^*}{3\hbar^2}$, and we find $\delta_N = n_U U^2 \frac{m^*}{3\hbar^2}$. In this case we have the relation $\delta_N = \frac{1}{2} \delta_G$. (iii) When the disorder potentials in the two layers are perfectly anticorrelated, $U_1 = -U_2$, in the limit $T \to T_c$, we have $\frac{1}{\tau_i} = n_U U^2 \frac{m^*}{3\hbar^2} + \frac{1}{\tau_i} = n_U U^2 \frac{m^*}{3\hbar^2}$. In this case we find $\delta_N = n_U U^2 \frac{m^*}{3\hbar^2}$, so that $\delta_N = \frac{1}{2} \delta_G$.

We summarize the magnitudes of the pair-breaking effect of disorder in the gapped and in the nematic phase under different interlayer disorder-correlation conditions in Table I. Irrespective of the interlayer correlations of disorder, the disorder suppression of $T_c$ is weaker in the nematic phase than in the gapped phase. Assuming $T_{c,0} < T^G_{c,0}$, we then find that the system can undergo a transition from the gapped phase to the nematic gapless phase by changing the strength of disorder, as shown in Fig. 1. Figure 1(a) shows $T_c$, obtained by solving Eq. (8), as a function of the intravalley disorder strength characterized by the dimensionless variable $d \equiv n_U U^2 m^*/(2\hbar^2 T_{c,0}^G)$, for the case of $T^N_{c,0}/T^G_{c,0} = 0.8$, in the gapped and the nematic phase under the three interlayer disorder-correlation conditions. Below a critical disorder strength the gapped phase is dominant while above it the nematic phase becomes preferable. The phase diagram calculated at various $d$ and $T^N_{c,0}/T^G_{c,0}$ in the case of $\delta_N = \delta_G/2$ is shown in Fig. 1(b).

If the dominant source of disorder is charge impurities, analogous to Eq. (10) we can then provide a quantitative estimate for the critical impurity density $n_{imp,c}$ above which the nematic phase is completely suppressed. We find

$$n_{imp,c} = A(3 \times 10^{10} \text{ cm}^{-2}) T^N_{c,0} \frac{\text{meV}}{\text{meV}},$$

(16)

where $A = 4/3, 2, 4$ depending on the interlayer correlation properties of disorder.

### B. Intervallel disorder scattering

In this section, we discuss the effect of intervalley disorder, i.e., $\tilde{W} \neq 0$. In this case the resulting valley-flip processes distinguish between states with different valley structure. In the following we consider the case in which the two types of disorder potential $\tilde{U}$ and $\tilde{W}$ are uncorrelated, and $|U_1| = |U_2| \neq U_1 = |W_1| = |W_2| \neq |W|$, and the density of intervalley scattering centers $n_w$ is the same in the two layers.

In the gapped phase, taking into account the presence of intervalley scattering, for the valley-independent states (LAF, QVH), the scattering rates in Eq. (7) become:

$$\frac{1}{\tau_i} = (n_U U^2 + n_w |W|^2) \frac{m^*}{3\hbar^2}, \quad \frac{1}{\tau_i} = 0,$$

indicating an enhancement on the pair-breaking effect characterized by $\delta_{G,\text{LAF}} = (n_U U^2 + n_w |W|^2) \frac{m^*}{3\hbar^2} = \delta_G(1 + \frac{n_w |W|^2}{n_U U^2})$. On the other hand, for the valley-polarized states (QAH, QSH), we obtain $\frac{1}{\tau_i} = n_U U^2 m^*/3\hbar^2$, indicating that the pair-breaking effect is unaltered since the influence of the intervalley disorder only introduces a non-pair-breaking component $\frac{1}{\tau_i}$.

Table II summarizes the effect of intervalley disorder on the different gapped states. Our results suggest that the valley-independent states (LAF, QVH) are more likely to appear in samples with very low disorder while the valley-polarized states (QAH, QSH) could survive at higher disorder concentrations.

For the nematic phase we find that, if $W_1$ and $W_2$ are uncorrelated, states with different valley structure are equally affected and therefore the intervalley disorder does not favor a specific valley structure.

### III. CONNECTION TO CURRENT EXPERIMENTS

Currently, two experimental groups have conducted comparative studies on samples with different disorder strengths: (i) The measurements presented in Ref. [23], performed on suspended and current annealed BLG devices, reveal two kinds of samples, B1 and B2. B2 samples are found to be gapped with vanishingly small conductance at the CNP in zero external fields, while B1 samples exhibit a small but finite conductance. The measurements show that B2 samples are cleaner than B1 samples. (ii) The most systematic study is done in Ref. [25]. In this work the authors investigate twenty-three high-quality suspended BLG devices and find that these samples, at low temperatures ($T < 10$ K) and zero external fields, fall into two groups: sixteen samples have a minimum conductivity of the order of 2 to 3 $e^2/h$, whereas seven samples are practically insulating with conductivity $\lesssim 0.4 e^2/h$. At the same time, the seven insulating samples are among the highest room-temperature-mobility samples, indicating a lower disorder strength in the insulating samples. Notice that the value of the minimum conductivity ($2$ to $3$ $e^2/h$) reported in Ref. [25] for the sixteen samples with lower mobility ($3 \times 10^8$ to $10^5$ cm$^2$ V$^{-1}$ s$^{-1}$) is quite smaller than the value of minimum conductivity expected for samples of this quality in the normal (nonordered) state of BLG [4]. It is then natural to expect that these sixteen samples, at low temperature, might be in a nematic or a gapped valley-polarized state and not in the normal state.

It is a possible scenario to interpret the results presented in Refs. [23,25] as suggesting that the cleanest samples are in a valley-independent gapped state that has no protected edge currents (insulating) and that the samples with lower mobility, due to higher disorder strength, are either in the nematic gapless phase or in a gapped valley-polarized state that has pro-
tected edge currents. This interpretation of the measurements of these comparative experimental studies is qualitatively consistent with our results that show that, as the strength of the nonmagnetic disorder increases, the valley-independent gapped states get suppressed more strongly and the nematic or the gapped valley-polarized states become favored. In addition, in the experiments presented in Refs. [20,21] it is estimated that the density of charge impurities in the sample that exhibits signatures of a broken-symmetry state is on the order of \(10^{10} \text{ cm}^{-2}\). This order of impurity density is consistent with our results, given that it is lower than the value that we obtain [Eqs. (10) and (16)] for the critical charge impurity density, above which \(T_c \rightarrow 0\), for both the gapped and the nematic phases, considering that, in the clean limit, \(T_c\) is on the order of a few meV.

The discussion above indicates that the effect of disorder described in our work should be directly relevant to current experiments on BLG, with some limitations. The experimental results presented in Refs. [23,25] clearly show that disorder plays an important role in determining the nature of the broken-symmetry state in BLG. Our work provides an insight on how nonmagnetic disorder might resolve the competition between different ordered states. Given the difficulty of probing experimentally the nature of the ordered phase, the strength of the disorder, and in particular the relative strength of intervalley and intravalley disorder, more work is needed to fully characterize the effect of the interplay between electron correlations and disorder in BLG.

IV. CONCLUSIONS

In conclusion, we have studied the effect of nonmagnetic disorder on the nature of bilayer graphene broken symmetry state that is expected to be established when the chemical potential is set at the charge-neutrality point even in the absence of external electric and magnetic fields. Current experiments have shown signatures suggesting that the broken-symmetry state could be either in a gapped phase or in a nematic gapless phase. For this reason we focused our analysis only on these two groups of ordered states, even though it has been shown theoretically that many other competing ordered states are possible [6–18].

We find that, in the presence of intravalley disorder, the resulting pair-breaking effects have different magnitude in the gapped and in the nematic phase: the transition temperature is suppressed more strongly in the gapped phase than in the nematic phase. Moreover, we find that in the nematic phase the pair-breaking effect of the disorder depends significantly on the interlayer correlation properties of the disorder: the pair-breaking effect is weaker in the uncorrelated case than in the perfectly correlated case, and it is the weakest for the case of perfectly anticrorelated disorder. We also find that the presence of intervalley disorder enhances the pair-breaking effect of disorder on the valley-independent gapped states but that it merely contributes a non-pair-breaking component to the valley-polarized gapped states.

Our results suggest that clean BLG might have a valley-independent gapped ground state (e.g., LAF), which does not have protected edge modes, but that small amounts of intervalley disorder can drive it into a valley-polarized gapped state with edge modes (e.g., QAH or QSH), and that intravalley disorder can drive it into a nematic state. The relation of our results to the current available experiments has been discussed. In addition, assuming charge impurities to be the dominant source of disorder, we provide a quantitative estimate of the critical impurity densities above which the gapped and the nematic order vanish, which can be tested in experiments.

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