Possible time-reversal-symmetry-breaking fermionic quadrupling condensate in twisted bilayer graphene

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We study the effective model for superconducting magic-angle twisted bilayer graphene beyond mean-field approximation by using Monte Carlo simulations. We consider the parameter regime where the low-temperature phase is a superconductor that spontaneously breaks time-reversal symmetry. When fluctuations are taken into account, it is shown that a fluctuations-induced phase with a fermion quadrupling order appears, where a different condensate, formed by four electrons, breaks time-reversal symmetry.

The recently discovered superconducting state which emerges in magic-angle twisted bilayer graphene exhibits a critical temperature that is exceptionally high compared to the Fermi energy [1–5]. This, and the fact that the system is two-dimensional, implies the presence of strong pairing fluctuations.

While superconductivity is a state of matter, which results from electron pairing, that is more than century-old, the presence of strong fluctuations suggest the tantalizing possibility that magic-angle twisted bilayer graphene can be an ideal system to realize different states of matter in the form of condensates of electronic quadruplets. In principle, the standard Bardeen-Cooper-Schrieffer theory does not allow fermionic quadrupling condensates. However, if the low-temperature regime of twisted bilayer graphene exhibits a superconducting ground state that breaks multiple symmetries, then, as we show below, it has the ideal ingredients for the formation of fluctuations-induced electron quadruplet states.

Multiple broken symmetries imply a multicomponent order parameter. Hence it is described by multiple complex fields of the form $|\Delta_i| e^{i\phi_i}$. Consider a system that is a two-dimensional multicomponent superconductor: at finite temperature, and for a finite magnetic-field penetration length, the only non-vanishing order parameter in the thermodynamic limit has to be constructed out of at least four fermionic fields [6–8]. This is based on the observation that composite superconducting vortices, which have identical phase winding in all components, have finite energy due to supercurrents screening effects. Therefore, a fluctuating two-component system is unstable to the proliferation of composite vortices that disorder the superconducting phase, while preserving the relative density or the phase difference between the components of the order parameter. The phase difference $\phi_i - \phi_j \propto \text{arccos} \frac{\Delta_i \Delta_j^*}{|\Delta_i||\Delta_j|}$ is an order parameter proportional to the product of two complex fields and hence represents four-fermion correlations. Various other realizations of four-fermion order were discussed in two-dimensional systems that exhibit multi-component superconductivity at zero temperature [3–9,15].

The recent microscopic study [10] derived an effective mean-field model for twisted bilayer graphene (TBG) near half filling of the valence band ($n = -2$). This model accounts for the six or twelve Van Hove points in order to capture the low-energy physics of the system [16]. In the case of the so-called six-patch model in [16], the superconducting phase can be effectively described by two superconducting (SC) order parameters $\Delta_1 = |\Delta_2| e^{i\phi_1}$ and $\Delta_2 = |\Delta_1| e^{i\phi_2}$, via the leading-order Landau functional:

$$V(|\Delta_1|,|\Delta_2|) = \alpha_1 (|\Delta_1|^2 + |\Delta_2|^2) + \beta_1 (|\Delta_1|^2 + |\Delta_2|^2)^2 + \beta_2 |\Delta_1^2 + \Delta_2^2|^2, \tag{1}$$

where $\alpha_1 \propto (T - T_c)$, with $T_c$ being the mean field critical temperature, $\beta_1 > 0$ and $\beta_1 + \beta_2 > 0$ for stability.

The free-energy potential Eq. (1) permits two different ground state manifolds, that are determined by the sign of the coupling $\beta_2$. For $\beta_2 > 0$, the ground state is a chiral superconductor that breaks time-reversal symmetry, while for $\beta_2 < 0$ the superconducting state develops a nematic order. Finally, for $\beta_2 = 0$, the potential exhibits an $SU(2)$ symmetry. According to the Mermin-Wagner theorem [17], two-dimensional systems with short-range interactions cannot spontaneously break a continuous symmetry at finite temperature. However, while for the case $SU(2)$ symmetry ($\beta_2 = 0$), the system does not exhibit any phase transition, the presence of a biquadratic term $\beta_2 \neq 0$, that explicitly breaks the $SU(2)$ symmetry into a $U(1) \times Z_2$ symmetry, allows for the emergence at low temperatures of an algebraic-ordered superconducting state that additionally breaks a $Z_2$ symmetry. Due to the atomically two-dimensional character of bilayer graphene, the magnetic field screening is negligible, hence one can neglect coupling to vector potential. In this case, while the system preserves a $U(1)$ symmetry at any finite temperature [17], it exhibits a SC phase transition belonging to the Berezinskii-Kosterlitz-Thouless universality class [18–20].

In the model [10], which differs notably from the phase-only models considered in [11–14], fluctuations in the density sector may dramatically impact the resulting phase diagram especially in the limit of small $\beta_2$ where the system approach the $SU(2)$ symmetry.
In this letter, we focus on the effect of fluctuations in the microscopic model \[16\] in the scenario \(\beta_2 > 0\). Starting with the free energy functional proposed in \[16\], we employ large-scale Monte Carlo simulations to obtain the phase diagram of the system beyond the mean-field approximation.

The Ginzburg-Landau free-energy density of the system reads:

\[
f = \sum_{i=1,2} \left[ \frac{1}{2} |\nabla \Delta_i|^2 + \alpha_1 |\Delta_i|^4 \right] + \beta_1 (|\Delta_1|^2 - |\Delta_2|^2)^2 + \beta_2 |\Delta_1|^2 + |\Delta_2|^2. \tag{2}\]

When coupled to a gauge field, this model only exhibits a quartic order in the thermodynamic limit \[6\]. When \(\rho \equiv \rho^K\), given by

\[
\rho^K = 1 - \frac{\alpha_1}{(\beta_1 + \beta_2)} \|\Delta_1\|^2, \tag{3}\]

given by \(K = \frac{\beta_2}{\pi^2 + \pi_2} > 0\). Next, by expressing the free energy in terms of phase-sum and phase-difference modes we obtain:

\[
f = \frac{1}{2} \rho^2 \left[ |\Delta_1|^2 |\nabla \phi_1|^2 + |\Delta_2|^2 |\nabla \phi_2|^2 \right] + \frac{|\Delta_1|^2 |\Delta_2|^2}{2\rho^2} \left[ |\nabla (\phi_1 - \phi_2)|^2 \right] + \frac{1}{2} \left[ |\nabla |\Delta_1||^2 + |\nabla |\Delta_2||^2 \right] + 2K |\Delta_1|^2 |\Delta_2|^2 [\cos(2(\phi_1 - \phi_2)) - 1], \tag{4}\]

with \(\rho^2 = |\Delta_1|^2 + |\Delta_2|^2 = 1\).

For finite values of \(K\), the ground state of the system shows an algebraic-ordered SC state that spontaneously breaks the \(Z_2\) symmetry. This results from the two-fold degeneracy of the phase difference \(\phi_{1,2} = \phi_1 - \phi_2 = \pm \pi/2\), due to the biquadratic Josephson term.

To obtain the phase diagram of the model \[1\], and in particular identify the presence of a quartic metal phase, it is necessary to use, as a function of the parameter \(K\), the two critical temperatures \(T_{BKT}\) and \(T_c(\Delta_2)\) for (i) \(T_{BKT} > T_c(\Delta_2)\), there arises a superconducting phase that preserves time-reversal symmetry; while for (ii) \(T_c(\Delta_2) > T_{BKT}\), a metallic state that breaks the time-reversal symmetry forms as a result of the condensation of fermion quadruplets \[11\] \[21\] \[23\]. The observation of a quadrupling-fermionic condensate was recently reported in the three-dimensional material Ba\(_{1-x}\)K\(_x\)Fe\(_2\)As\(_2\) \[21\].

Beyond the mean-field approximation, the physics of the system, and therefore its phase diagram, is governed by the proliferation of topological phase excitations. These can be elementary vortex excitations, resulting from a phase winding in each condensate individually; composite vortices, resulting from the phase winding of both condensates around the same core; and domain walls separating regions with opposite phase-differences. The elementary vortices \((\Delta \phi_1 = \pm 2\pi, \Delta \phi_2 = 0) \equiv (\pm 1, 0)\) or \((\Delta \phi_1 = 0, \Delta \phi_2 = \pm 2\pi) \equiv (0, \pm 1)\) have a phase winding in the inter-component phase difference and hence emit a domain wall. Consequently their proliferation restores the \(Z_2\) symmetry and simultaneously destroys the superconducting state leading to the BKT superfluid-stiffness jump to zero at the critical point \[20\]. On the other hand, the proliferation of composite vortices of the kind \((1, 1)\) can only affect the superconducting sector, leaving the \(Z_2\) symmetry broken. Likewise, the proliferation of domain-wall excitations alone can only restore the \(Z_2\) symmetry leaving the superfluid stiffness associated with the SC phase finite. The restoration of short-range order upon heating is thus determined by the interplay of different topological defects, which is a highly nontrivial problem that cannot be settled analytically.

In this work, we address this phase transition via large-scale Monte Carlo simulations of the two-dimensional model \[4\]. The discrete Hamiltonian reads:

\[
H = - \sum_{i,\mu} \sum_{\alpha=1,2} |\Delta_{\alpha,i}| |\Delta_{\alpha,i+\mu}| \cos(\phi_{\alpha,i+i} - \phi_{\alpha,i}) + K |\Delta_{1,i}|^2 |\Delta_{2,i}|^2 [\cos(2(\phi_{1,i} - \phi_{2,i})) - 1], \tag{5}\]

where \(\mu = \hat{x}, \hat{y}\) and \(|\Delta_{1,i}|^2 + |\Delta_{2,i}|^2 = 1 \forall i \in [0, L \times L]\). Further details of the numerical simulations are discussed in the Supplementary Information.

The BKT superconducting transition is associated with the emergence of a finite stiffness of the phase-sum. Within the Ginzburg-Landau model Eq. \[3\], this can be assessed by computing the helicity-modulus sum \(Y^\mu_\delta\), defined as the linear response of the system to an infinitesimal twist of the two phase condensates along \(\mu\):}

\[
Y^\mu_\delta = \left. \frac{1}{L^2} \frac{\partial^2 F(\{\phi_i\})}{\partial \delta^\mu} \right|_{\delta^\nu = 0} = \sum_i Y^\mu_\delta + 2Y^\mu_\delta, \tag{6}\]
where:

$$\Upsilon_+ = \frac{1}{L^2} \left[ \frac{\partial^2 H}{\partial \delta_{\mu,i}^2} - \beta \left( \frac{\partial H}{\partial \delta_{\mu,i}} \right)^2 \right]_{\delta=0} ,$$

$$\Upsilon_{12} = -\frac{\beta}{N} \left[ \frac{\partial H}{\partial \delta_{\mu,1} \delta_{\mu,2}} - \beta \left( \frac{\partial H}{\partial \delta_{\mu,1}} \right) \left( \frac{\partial H}{\partial \delta_{\mu,2}} \right) \right]_{\delta_{\mu,12}=0} .$$

Here, $L$ is the linear size of the two-dimensional system. The expectation value $\langle \ldots \rangle$ is the thermal average, evaluated stochastically by Monte Carlo. In what follows, we will simply write: $\Upsilon_+ \equiv \Upsilon_+^*$.

Ordinary $U(1)$ systems in two dimensions exhibit a topological phase transition driven by the unbinding of vortex-antivortex pairs [19, 20, 27], which becomes entropically favorable at a finite temperature $T_{BKT}$. The proliferation of free vortices leads to a discontinuous vanishing of the phase stiffness, that drops to zero at $T_{BKT}$ according to the Kosterlitz-Nelson universal relation [28].

When a system undergoes a BKT phase transition [19, 20, 27], the critical point can be located by finite-size scaling of the quantity [25]:

$$\Upsilon_+(\infty, T_{BKT}) = \Upsilon_+(L, T_{BKT}) \left( \frac{1}{1 + (2 \log(L/L_0))^{-1}} \right) ,$$

where $L_0$ is a free parameter giving the best crossing point at finite temperature (see also Supplementary information and Supplementary Fig. 1). For $K = 5$, the best crossing point is obtained for $L_0 = 3$, as shown in Fig. 1(b). Varying $K$, the value of $L_0$ varies as well. In particular, we find that $L_0$ increases by decreasing $K$ (see Supplementary Fig. 2 and Supplementary Fig. 3), leading to very pronounced finite-size effects at small $K$. This finding stems from the multi-component nature of the system. Indeed, in contrast to the single-component case, the BKT transition is in this case driven by the proliferation of free composite vortices, resulting from the unbinding of a pair formed by a (1,1) and a (−1,−1) vortex. For large values of $K$, the superconducting phases of the two condensates are essentially locked, and the model (3) can effectively be described by a single component.

In this limit, the two elementary vortices $\pm(1,0)$ and $\pm(0,1)$ that constitute $\pm(1,1)$ composite vortices are tightly bound. However, for smaller values of $K$, this is no longer the case. Moreover, the density-density interaction promotes the separation of the composite vortices into their elementary constituents since in the limit $K \to 0$ the model approaches the $SU(2)$ symmetry where the composite vortices are unstable. The finite size of these composite vortices can therefore increase the finite-size effects of the whole system resulting in a larger value of $L_0$.

To assess the $Z_2$ phase transition, we define an effective Ising order parameter $m$, related to the two possible values of $\phi_{1,2} \in [-\pi; \pi]$ via:

$$\begin{cases} m = +1 & \phi_{1,2} \geq 0 \\ m = -1 & \phi_{1,2} < 0 . \end{cases}$$

Finally, we extract the $Z_2$ critical temperature by means of a finite-size crossing analysis of the Binder cumulant $U$ of $m$:

$$U = \frac{\langle m^4 \rangle}{3 \langle m^2 \rangle^2} .$$

In the thermodynamic limit, $U$ tends to 1 in the high-temperature phase and to 1/3 in the low-temperature limit. The resulting crossing point for the case $K = 5$ is shown in Fig. 1(a).
FIG. 2: Phase diagram of the model Eq. (3) as a function of the coupling $K$. For any finite value of $K$, the BKT and $Z_2$ transitions are found to be separated, with $\beta_{BKT} > \beta_c(Z_2)$. In the inset, the size of the observed splitting $\Delta \beta_c = \beta_{BKT} - \beta_c(Z_2)$ is reported as function of $K$. The largest splitting is found for smaller values of $K$, while in the limit of $K \to \infty$ it saturates to a finite value.

The phase diagram obtained via this numerical study is shown in Fig. 2. Details on the finite-size scaling of the two critical temperatures can be found in the Supplementary Information (Supplementary Fig. 4-6). Our results reveal that for any finite value of $K$ considered, the system has a fermionic quadrupling state that breaks time-reversal symmetry. The range of temperatures where this phase appears (see the inset of Fig. 2) is larger for smaller values of $K$ and saturates to a finite value in the limit $K \to \infty$.

In conclusion, we find that the model derived from twisted bilayer graphene [16] hosts a fermion quadrupling phase above the superconducting phase. This phase is significantly larger and more robust than previously considered scenarios in different models [11, 21, 22]. Importantly, it extends to all finite values of the coupling parameter $K$. Note that, in contrast to conventional multiband models, in the limit of a vanishing intercomponent Josephson coupling $K$, the symmetry of the system derived in [16] is $SU(2)$, rather than $U(1) \times U(1)$ (as is the case for $s + is$ superconductors). In two dimensions, $SU(2)$-symmetric systems exhibit no long-range or quasi-long range order. Hence, approaching this point, density-fluctuations become stronger and, as our simulations show, the critical temperature of the BKT-transition shifts relative to the $Z_2$ critical temperature.

Obtaining a significant fermion quadrupling phase in the case when $U(1) \times U(1)$ symmetry is explicitly broken to $U(1) \times Z_2$ generally requires a strong symmetry-breaking Josephson term [11]. By contrast, in the TBG-model considered in this work, the quadrupling phase remains large, even if the term that breaks $Z_2$ symmetry is small. This fact indicates that magic-angle twisted bilayer graphene can be an especially promising platform for realizing and observing the fermion quadrupling order. Notably, this order can be further enhanced by applying external magnetic fields [17, 24]. The fermion quadrupling state can be identified via a combination of thermal and electrical transport measurements, analogous to those performed in [24].

The effective model of the $Z_2$ quadrupling state [29] suggests that signatures of a $Z_2$ broken symmetry above the critical temperature can be detected via magnetic probes. Skyrmion excitations or spontaneous magnetic fields can indeed appear in the presence of local strain, obtained by imposing local pressure or by local heating [30, 31] in combination with local magnetic probe.

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SUPPLEMENTARY INFORMATION

S1. DETAILS OF MONTE CARLO SIMULATIONS

We have performed Monte Carlo (MC) simulations of the discrete model:

\[
H = - \sum_{i,\mu} \sum_{\alpha=1,2} |\Delta_{\alpha,i}| |\Delta_{\alpha,i+\mu}| \cos (\phi_{\alpha,i+\mu} - \phi_{\alpha,i}) + \sum_i K |\Delta_{1,i}|^2 |\Delta_{2,i}|^2 \cos (2(\phi_{1,i} - \phi_{2,i})) - 1, \tag{12}
\]

where \( \mu = \hat{x}, \hat{y} \), locally updating via Metropolis-Hastings algorithm both the two phase fields \( \phi_1, \phi_2 \in [0,2\pi) \) and two amplitude fields \( |\Delta_1|, |\Delta_2| \) with the constraint \( |\Delta_1|^2 + |\Delta_2|^2 \).

A single MC step consists of the Metropolis sweeps of the whole \( L \times L \) lattice fields. To speed-up the thermalization, we also implemented a parallel tempering algorithm, allowing swap of field configurations between neighbouring temperatures. Typically, we propose one set of swap after 32 MC steps. For most of the numerical simulations, we performed a total of \( 3 \times 10^5 \) Monte Carlo steps, discarding the transient time occurring within the first 60000 steps. For the simulation performed in the limit of large \( K \) we implemented a cluster update, similar to the Wolff algorithm for the Ising model, to prevent the system from getting stuck in metastable states.

S2. ASSESSING THE BEREZINSKII-KOSTERLITZ-THOULESS TRANSITION

The BKT critical point can be located by finite-size scaling of the quantity of the helicity modulus sum [1]

\[
\frac{\pi \Upsilon_+(L,T_{BKT})}{2T_{BKT}} = 1 + \frac{1}{2 \ln(L/L_0)}; \tag{13}
\]

that can be rewritten as:

\[
\Upsilon_+(\infty,T_{BKT}) = \frac{\Upsilon_+(L,T_{BKT})}{1 + (2\log(L/L_0))^{-1}}. \tag{14}
\]

Typically \( L_0 \) is treated as a free parameter giving the best crossing point at finite temperature. However, it is in principle possible to extrapolate \( L_0 \) using the BKT scaling itself. In the present analysis we proceeded as follows. We rewrite Eq.(13) as:

\[
\ln(L) - \frac{1}{2(x_L(T_{BKT}) - 1)} = \ln(L_0), \tag{15}
\]

where \( x_L(T) = \frac{\pi \Upsilon_+(L,T)}{2T} \). Thus, by looking at the crossing point of the function:

\[
f(L,T) = \ln(L) - \frac{1}{2(x_L(T) - 1)}, \tag{16}
\]

plotted as function of the temperature for different values of \( L \), we can directly obtain the value of \( L_0 \). We use this procedure to obtain the value of \( L_0 \), as shown for the case \( K = 5 \) in Fig.4.

The values of \( L_0 \) extracted as function of \( K \) are shown in Fig.4. In Fig.5, we report the BKT crossing points for all values of \( K \) considered.
FIG. 3: Left panel: temperature dependence of the helicity modulus sum rescaled according to Eq. (14) for different values of the linear size $L$. The value of $L_0$ has been obtained from the crossing point of the function $f(L, T) = \ln(L) - \frac{1}{2(\alpha L(T) - 1)}$ shown in the right panel.

FIG. 4: Value of the parameter $L_0$ as function of the Josephson coupling $K$. By decreasing $K$, $L_0$ increases as a result of increasing finite-size effects due to the size of the composite topological vortices driving the BKT phase transition.
FIG. 5: Temperature dependence of the helicity-modulus sum $\Upsilon_\alpha$ renormalized according to the BKT scaling for different values of the linear size $L$ and the coupling $K$. 
S3. FINITE-SIZE ANALYSIS OF THE SPLITTING BETWEEN THE BKT AND \(Z_2\) PHASE TRANSITIONS

FIG. 6: Finite-size scaling of the inverse of the two critical temperatures \(\beta_{BKT}\) and \(\beta_c(Z_2)\), for: (a) \(K = 0.2\); (b) \(K = 0.5\). To extrapolate the thermodynamic limit of the two critical temperatures, we proceed as follows. When the linear fitting of \(\beta_c\) as function of \((L_iL_{i+1})^{-1/2}\) returns a value of \(R^2\) larger than a threshold we fixed to 0.6, we take the value of the intercept with the y axis as an estimate for the thermodynamic value of \(\beta_c(L \rightarrow \infty)\), in the other cases we take the mean value of \(\beta_c(L_iL_{i+1})^{-1/2}\) estimating its statistical error by the standard error-propagation relations. For the cases where \(R^2 > 0.6\) the linear fit is shown as a dashed line. The values of \(L_i\) used are for each \(K\) larger than the corresponding \(L_0\). More specifically: for \(K = 0.2\) we used \(L = 80, 96, 128, 160, 192, 256\); for \(K = 0.5\) we used \(L = 64, 80, 96, 128, 160, 192, 256\).
FIG. 7: Finite-size scaling of the inverse of the two critical temperatures $\beta_{BKT}$ and $\beta_c(Z_2)$, for: (a) $K = 1$; (b) $K = 5$; (c) $K = 10$. To extrapolate the thermodynamic limit of the two critical temperatures, we proceed as follows. When the linear fitting of $\beta_c$ as function of $(L_i L_{i+1})^{-1/2}$ returns a value of $R^2$ larger than a threshold we fixed to 0.6, we take the value of the intercept with the y axis as an estimate for the thermodynamic value of $\beta_c(L \to \infty)$, in the other cases we take the mean value of $\beta_c(L_i L_{i+1})^{-1/2}$ estimating its statistical error by the standard error-propagation relations. For the cases where $R^2 > 0.6$ the linear fit is shown as a dashed line. The values of $L_i$ used are for each $K$ larger than the corresponding $L_0$. More specifically: for $K = 1$ we used $L = 40, 48, 64, 80, 96, 128, 160, 192, 256$; for $K = 5$ we used $L = 12, 16, 20, 24, 32, 40, 48, 64, 96, 128$; for $K = 10$ we used $L = 10, 12, 16, 20, 24, 32, 40, 48, 64, 96, 128$. 
FIG. 8: Finite-size scaling of the inverse of the two critical temperatures $\beta_{BKT}$ and $\beta_c(Z_2)$, for: (a) $K = 15$; (b) $K = 20$; (c) $K = 50$. To extrapolate the thermodynamic limit of the two critical temperatures, we proceed as follows. When the linear fitting of $\beta_c$ as function of $(L_i L_{i+1})^{-1/2}$ returns a value of $R^2$ larger than a threshold we fixed to 0.6, we take the value of the intercept with the y axis as an estimate for the thermodynamic value of $\beta_c(L \to \infty)$, in the other cases we take the mean value of $\beta_c(L_i L_{i+1})^{-1/2}$ estimating its statistical error by the standard error-propagation relations. For the cases where $R^2 > 0.6$ the linear fit is shown as a dashed line. The values of $L_i$ used are for each $K$ larger than the corresponding $L_0$. More specifically: for $K = 20$ and for $K = 15$ we used $L = 10, 12, 16, 20, 24, 32, 40, 48, 64, 128$; for $K = 50$ we used $L = 8, 10, 12, 16, 20, 24, 32, 40, 48, 64, 128$.
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