Time-reversal versus chiral symmetry breaking in twisted bilayer graphene

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By applying a self-consistent Hartree-Fock approximation, we show that the mechanism of dynamical symmetry breaking can account for the insulating phase that develops around the charge neutrality point of twisted bilayer graphene at the magic angle. At strong and intermediate coupling of the Coulomb interaction, the opening of a gap between the lowest-energy valence and conduction bands proceeds preferentially through the breakdown of chiral symmetry. This effect is mainly driven by the extended component of the interaction. When this is under very strong dielectric screening, however, we enter a regime where the on-site Coulomb repulsion becomes dominant, leading to the prevalence of time-reversal and parity symmetry breaking in a weak-coupling window of the phase diagram. Moreover, when the long-range tail of the Coulomb interaction is not screened, we see the appearance of another dominant pattern, which is characterized by breaking the time-reversal invariance but preserving parity. This gives rise to the superposition of two different effects, namely the opening of a gap at the Dirac cones and the splitting of the degeneracy of the low-energy bands at the $K'$ points of the Moiré Brillouin zone.

Introduction. The discovery of superconductivity\textsuperscript{1} next to insulating phases\textsuperscript{2} in twisted bilayer graphene at small twist angle has opened a new era in the investigation of strong electron correlations in 2D materials.\textsuperscript{3–34} The strong interaction effects arise from the progressive narrowing of the first valence and conduction bands, with the bandwidth reaching a minimum of the order of $\sim 1$ meV at the so-called magic angle. The strong correlation effects appear at integer fillings of the superlattice of the twisted bilayer, suggesting that the large effective interaction strength may be the driving force behind them. This effect has recently been verified in local probe experiments.\textsuperscript{35–38}

A remarkable effect is the insulating behavior observed at the charge neutrality point of the twisted bilayer.\textsuperscript{7} This may be somewhat surprising since, in the undoped carbon material, the Fermi level is placed at the vertices of the Dirac cones characterizing the low-energy dispersion in the Moiré Brillouin zone. As long as the density of states vanishes at that filling level, it is pertinent to ask about the mechanism responsible for the opening of a gap at the charge neutrality point.

This discussion recalls the highly debated question about the dynamical breakdown of chiral symmetry in Dirac-like systems.\textsuperscript{39–46} That kind of electronic instability has not been observed in graphene, despite the large nominal coupling of the Coulomb potential in the carbon layer.\textsuperscript{47–49} Nevertheless, in bilayer graphene correlated insulating states were seen\textsuperscript{50,51} and in the case of the twisted bilayer, the relative strength of the Coulomb interaction is further enhanced at the magic angle, making plausible that the critical coupling for the opening of a gap at the Dirac cones may be surpassed.

In this paper, we study the effects of the Coulomb interaction at the charge neutrality point of twisted bilayer graphene near the magic angle, with the aim of discerning whether dynamical symmetry breaking takes place under different screening conditions. Our analysis focuses on a twisted bilayer corresponding to $i = 28$ (twist angle $\theta \approx 1.16^\circ$) in the sequence of commensurate superlattices with twist angle $\theta_i = \arccos((3i^2 + 3i + 0.5)/(3i^2 + 3i + 1))$.\textsuperscript{52–54} This allows us to investigate a relevant representative in which the combined bandwidth of the first valence and conduction bands has a small magnitude $W \sim 1$ meV, but still larger than the value of the Coulomb repulsion in the Moiré superlattice.

We adopt a tight-binding approach to make a real space description of the system. In this framework, we resort to a Hartree-Fock approximation in order to assess the effects of the Coulomb interaction.\textsuperscript{36,38,55–57} The self-consistent method is well suited to compute the condensates signaling the breakdown of symmetry. The twisted bilayer is a complex system where we find the interplay between different degrees of freedom (two valleys, two layers), leading to a number of order parameters. The resulting picture is that several transitions take place between different phases as one modifies the strength of the Coulomb interaction. This mainly favors the breakdown of chiral and time-reversal symmetry, whose order parameters tend to compete along the phase diagram and become alternatively dominant at different regimes of the coupling strength.

Hartree-Fock approximation. Our starting point to model twisted bilayer graphene is a tight-binding approach in which we consider the extended hopping between all the carbon atoms in the two layers. The Hamiltonian $H_0$ of the non-interacting theory can be written in terms of creation (annihilation) operators $a_{i\sigma}^\dagger$ ($a_{i\sigma}$) for electrons at each site $i$ with spin $\sigma$ as\textsuperscript{58,59}

\begin{align}
H_0 &= -\sum_{\langle i,j \rangle} t_\parallel (r_i - r_j) \left( a_{i\sigma}^\dagger a_{j\sigma} + \text{h.c.} \right) \\
&\quad -\sum_{\langle i,j \rangle} t_\perp (r_i - r_j) \left( a_{i\sigma}^\dagger a_{j\sigma} + \text{h.c.} \right),
\end{align}

where the sum over the brackets $\langle \ldots \rangle$ runs over pairs of
atoms in the same layer, whereas the sum over the curved brackets \( \ldots \) runs over pairs with atoms belonging to different layers. \( t_0^\parallel (r) \) and \( t_0^\perp (r) \) are hopping matrix elements which have an exponential decay with the distance \(|r|\) between carbon atoms.\(^{60,61}\)

On top of that noninteracting model, we incorporate the effect of the Coulomb potential \( v(r) \). Then we add to \( H_0 \) the interaction hamiltonian

\[
H_{\text{int}} = \sum_{i,j} a_{i\sigma}^\dagger v(r_i - r_j) a_{j\sigma} a_{j\sigma}^\dagger \, .
\]

(2)

In general, one can discriminate between two different components of the interaction. One of them arises from the long-range part of the Coulomb potential, which has a \( 1/|r| \) decay but may be screened by the environment and affected also by internal screening. The other component corresponds to the on-site repulsion for electrons at the same carbon atom, which is rather insensitive to screening by the environment.

We deal next with the many-body problem, with the aim of computing the electron propagator \( G \) of the interacting theory. This can be obtained in terms of the propagator \( G_0 \) of the noninteracting system and the electron self-energy \( \Sigma \) according to the Dyson equation

\[
G^{-1} = G_0^{-1} - \Sigma \, .
\]

(3)

In what follows we are going to be interested in time-independent observables, which means that we can simplify the discussion by taking the zero-frequency (static) limit of the equations. In that limit, the free propagator \( G_0 \) is given in real space by the inverse of the matrix representation of the operator \( H_0 \), that is,

\[
(G_0)_{ij} = -(H_0^{-1})_{ij} \, .
\]

(4)

In terms of the eigenvalues \( \varepsilon_a^0 \) and eigenvectors \( \phi^0_a(r_i) \) of the noninteracting hamiltonian, we have therefore in the static limit

\[
(G_0)_{ij} = -\sum_a \frac{1}{\varepsilon_a^0} \phi^0_a(r_i) \phi^0_a(r_j)^* \, .
\]

(5)

The Hartree-Fock approximation amounts to assume that the full propagator can be also represented in terms of a set of modified eigenvalues \( \varepsilon_a \) and eigenvectors \( \phi_a(r_i) \):

\[
(G)_{ij} = -\sum_a \frac{1}{\varepsilon_a} \phi_a(r_i) \phi_a(r_j)^* \, .
\]

(6)

One can check (in the full time-dependent theory) that the assumption in Eq. (6) implies the representation of the self-energy matrix

\[
\Sigma_{ij} = 2 t_{ij} v(r_i - r_j) \sum_a |\phi_a(r_i)|^2
- v(r_i - r_j) \sum_a |\phi_a(r_i)\phi_a(r_j)^*| \, ,
\]

(7)

where the prime means that the sum is to be only carried over the occupied levels. The problem reverts to the fact that (3) and (7) are a set of self-consistent equations, as the self-energy is built from the same eigenvectors one seeks to find. In the present context, the problem demands the diagonalization of matrices with large dimension according to the number of atoms in the unit cell of the twisted bilayer. In practice, one can devise a recursive approximation to \( G \) in which good convergence is achieved by building the self-energy at each step with the eigenvectors obtained in the previous iteration.

Order parameters. In Fig. 1, we schematically show the dominant order parameters as they arise in single layer graphene, namely the inversion symmetry breaking giving rise to a Dirac mass\(^{62}\) and the time-reversal symmetry breaking giving rise to the Haldane mass\(^{63}\). We will now extend this analysis within the Hartree-Fock approximation to twisted bilayer graphene. Since we will not discuss possible topological phases, we set all prefactors in the following to one.

The Hartree-Fock approximation is a convenient approach to study symmetry breaking in the twisted bilayer, as the different order parameters can be expressed using the eigenvectors obtained in the self-consistent resolution. In this respect, we are going to see that the different symmetry breaking patterns can be cast in terms of the matrix elements

\[
h_{ij} = \sum_a \phi_a(r_i) \phi_a(r_j)^* \, ,
\]

(8)

where the prime means again that the sum is over the filled levels.

We study here the system with the Fermi level placed at the charge neutrality point, paying attention to the possibility that the Dirac nodes at the \( K \) points of the Moiré Brillouin zone may be destabilized due to inter-
action effects. A well-known mechanism to give mass to Dirac fermions is to induce a charge asymmetry between the two triangular sublattices of the honeycomb lattice, breaking the chiral symmetry. In the present case, we have sublattices \( A_1, B_1 \) for the top carbon layer and \( A_2, B_2 \) for the bottom layer. Thus, we may write two different order parameters for the breakdown of chiral symmetry, being respectively even and odd under the exchange of the two layers, as

\[
C_{\pm} = \sum_{i \in A_1} h_{ii} - \sum_{i \in B_1} h_{ii} \pm \left( \sum_{i \in A_2} h_{ii} - \sum_{i \in B_2} h_{ii} \right). \tag{9}
\]

Furthermore, another way to open a gap for Dirac fermions is to generate the so-called Haldane mass, which is characterized in the honeycomb lattice by complex hopping between next-to-nearest neighbors. We may label for instance as \( i_1, i_2 \) and \( i_3 \) the three nearest neighbors of each atom \( i \) of the twisted bilayer, with the order of the labels corresponding to clockwise orientation. Then, as in the case of chiral symmetry breaking, we have two different order parameters for the breakdown of time-reversal invariance (and parity), being respectively even and odd under the exchange of the two layers. They are given by the two choices

\[
P_{\pm} = \text{Im} \left( \sum_{i \in A_1} (h_{i_1i_2} h_{i_2i_3} h_{i_3i_1})^{1/3} + \sum_{i \in B_1} (h_{i_1i_2} h_{i_2i_3} h_{i_3i_1})^{1/3} \right)
\[
\pm \sum_{i \in A_2} (h_{i_1i_2} h_{i_2i_3} h_{i_3i_1})^{1/3} \pm \sum_{i \in B_2} (h_{i_1i_2} h_{i_2i_3} h_{i_3i_1})^{1/3} \right), \tag{10}
\]

The imaginary part of the product of the three matrix elements in (10) gives a measure of the flux through the twisted bilayer, providing a signature of the breakdown of time-reversal invariance. In the Haldane phase, that implies also the breakdown of parity, since the orientation of the labels is supposed to be the same around atoms in different sublattices of the honeycomb lattice.

We may envisage however the possibility that the direction of the flux may be the opposite in the two sublattices. That pattern corresponds to the breakdown of time-reversal symmetry, but preserving the parity invariance under the exchange of the sublattices. The order parameters for that phase may be even or odd with respect to the exchange of the two carbon layers, which translates into the two possibilities

\[
S_{\pm} = \text{Im} \left( \sum_{i \in A_1} (h_{i_1i_2} h_{i_2i_3} h_{i_3i_1})^{1/3} - \sum_{i \in B_1} (h_{i_1i_2} h_{i_2i_3} h_{i_3i_1})^{1/3} \right)
\]

\[
\pm \sum_{i \in A_2} (h_{i_1i_2} h_{i_2i_3} h_{i_3i_1})^{1/3} \pm \sum_{i \in B_2} (h_{i_1i_2} h_{i_2i_3} h_{i_3i_1})^{1/3} \right), \tag{11}
\]

It can be seen that the development of nonvanishing order parameters \( S_{\pm} \) leads to the splitting of the low-energy bands at the \( K \) points of the Moiré Brillouin zone.

This is a consequence of the fact that, in the low-energy theory of Dirac fermions, such a breakdown of symmetry proceeds with the dynamical generation of a diagonal term in pseudospin space, rather than with a term proportional to \( \sigma_z \). The diagonal term still preserves the Dirac nodes at the \( K \) points, but leading to a mismatch in the shift of the bands in the two valleys of the twisted bilayer.

Besides, we have to include the possibility of having an spontaneous imbalance of charge in the two carbon layers as a result of interaction effects. The order parameter for such a symmetry breaking is

\[
L = \sum_{i \in A_1} h_{ii} \pm \sum_{i \in B_1} h_{ii} - \sum_{i \in A_2} h_{ii} - \sum_{i \in B_2} h_{ii}. \tag{12}
\]

The main effect of the charge imbalance is to create a bias in the potential of the two carbon layers, leading to gap opening in the Dirac cones at the corners of the Moiré Brillouin zone.

This completes the list of the symmetry breaking patterns described from a real space point of view, and which have a more drastic effect giving rise to the destabilization of the Dirac nodes at charge neutrality. As long as we are dealing with spin-independent interactions, based on the Coulomb potential, it is justified to restrict here our analysis to those order parameters not involving the spin degree of freedom. This is also motivated by the experimental observations of twisted bilayer graphene at the charge neutrality point, where the main electronic instability (the transition to an insulating state) only affects to the charge degrees of freedom.

**Screened Coulomb interaction.** We first consider a form of the Coulomb potential which is adapted to the case where twisted bilayer graphene is surrounded by top and bottom metallic gates. We start with the unscreened Coulomb potential

\[
v_0(r) = \frac{e^2}{4\pi \epsilon} \frac{1}{r}, \tag{13}
\]

where \( \epsilon \) denotes the dielectric constant of the surrounding (non-metallic) medium. In the presence of a gate at distance \( d = \xi/2 \), the electrostatic energy of two electrons lying in a plane parallel to the electric gate and being separated by a distance \( r \) is given by

\[
v(r) = \frac{e^2}{4\pi \epsilon} \left( \frac{1}{r} - \frac{1}{\sqrt{r^2 + \xi^2}} \right). \tag{14}
\]

In the presence of an additional opposite gate also at distance \( d = \xi/2 \), and again using the image-charge technique, one obtains for the electrostatic energy

\[
v(r) = \frac{e^2}{4\pi \epsilon} \sum_{n=\infty}^{\infty} \frac{(-1)^n}{\sqrt{r^2 + n^2 \xi^2}}
\]

\[
= \frac{e^2}{4\pi \epsilon} \frac{2\sqrt{2} e^{-\pi r/\xi}}{\xi \sqrt{r/\xi}}. \tag{16}
\]
Assuming a Coulomb interaction mediated by the potential \((16)\) with \(\xi = 10\) nm, we have applied the Hartree-Fock approximation to a twisted graphene bilayer corresponding to \(i = 28\) (twist angle \(\theta \approx 1.16^\circ\)) in the sequence of commensurate superlattices. In that approach, the potential has to be still complemented with the value of the Coulomb repulsion for electrons at the same carbon atom, as the expression \((16)\) is ill-defined for \(r = 0\). We have taken such an on-site Coulomb repulsion equal to 4 eV, that is, we have regularized the Coulomb potential with the prescription

\[
v(r)|_{r=0} = 4 \text{ eV}.
\]  

(17)

Given that the number of carbon atoms in the Moiré unit cell for \(i = 28\) is \(N \sim 10000\), the magnitude of the effective Hubbard repulsion in the Moiré superlattice turns out to be \(v(0)/N \lesssim 1\) meV. This is below the magnitude of the bandwidth for the first valence and conduction bands of the chosen twisted bilayer, which is \(W \sim 1\) meV.

We have mapped the different symmetry breaking patterns as the strength of the Coulomb potential is modified, which may be achieved in practice by changing the dielectric constant \(\epsilon\). The evolution of the main order parameters can be seen in Fig. 2. There we observe that the order parameter \(C_+\) for the breakdown of chiral symmetry becomes dominant throughout all the range of strong and intermediate coupling of the Coulomb potential. Only when \(C_+\) starts to fade away, as the coupling strength is much reduced (large \(\epsilon\)), we find the onset of the order parameter \(P_+\), which develops a strong peak marking the transition to a phase with the breakdown of parity and time-reversal invariance. Concomitant with the two mentioned symmetry breaking patterns, we find the development of a gap at the \(K\) point of the Moiré Brillouin zone, represented also in Fig. 2.

We note that the behavior of the order parameter \(C_+\) at weak coupling is consistent with that expected for a Kosterlitz-Thouless transition, with a decay which is first relatively fast, and then slower close to the critical point. The evolution of \(P_+\) is rather different and it reflects the competition with \(C_+\) at the onset of chiral symmetry breaking. The fact that a strong peak of \(P_+\) appears at weak coupling (large \(\epsilon\)) makes plausible that it may arise from the on-site component of the Coulomb interaction. This is not affected by dielectric screening in our approach, making possible that a strong time-reversal symmetry breakdown becomes dominant when the Coulomb potential is already much suppressed by large values of \(\epsilon\), as seen in Fig. 2.

**Long-range Coulomb interaction.** It is also relevant to study the breakdown of symmetry when the twisted bilayer is not surrounded by metallic gates. This instance is interesting since it can give a measure of the strong effects expected from the long-range Coulomb interaction. We will then assume in this case that the Coulomb potential is given by

\[
v(r) = \frac{e^2}{4\pi\epsilon r},
\]

(18)

with the screening effects just encoded in the dielectric constant \(\epsilon\). As in the previous section, we also assume that the interaction at \(r = 0\) is regularized to coincide with the on-site Coulomb repulsion of the electrons at the same carbon atom, which we take equal to 4 eV as in Eq. (17).

In this case, the symmetry breaking patterns which play a dominant role to open a gap at the Dirac cones correspond again to the order parameters \(C_+\) and \(P_+\). Their competition can be observed in Fig. 3, in which we plot the different behaviors as the coupling of the Coulomb potential is modified by changing \(\epsilon\). There are similarities with the evolution shown in Fig. 2, like the opening of a Kosterlitz-Thouless transition for chiral symmetry breaking. This gives the dominant contribution to the gap opened in the Dirac cones at intermediate and strong coupling, although such a gap is now reinforced by the coincident development of time-reversal symmetry breaking in the latter regime. Moreover, we find again the strong peak of \(P_+\) at weak coupling, before the order parameter \(C_+\) takes over, which can be attributed to the unscreened on-site component of the Coulomb interaction.

Anyhow, the most remarkable feature observed in Fig. 3 is the prevalence of the pattern breaking time-reversal symmetry but preserving parity, characterized by the order parameter \(S_+\). This starts to grow under the dome of \(C_+\), but reaches much higher values than all the other order parameters in the strong coupling regime. As mentioned before, a nonvanishing value of \(S_+\) does not contribute to open a gap at the Dirac cones, which means
that the gap plotted in the figure is only an effect of the breakdown of symmetry characterized by $C_+$ and $P_+$.

The lowest-energy valence and conduction bands proceed and intermediate coupling, the opening of a gap between twisted bilayer graphene near the magic angle. At strong ing phase that develops at the charge neutrality point of namical symmetry breaking can account for the insulat-

$\gap (in eV, magnified by the factor shown in the figure).$

$\gap = 28$ at the charge neutrality point, as function of the

$\theta (=1.16^\circ)$ long-range

Summary. We have shown that the mechanism of dy-

namical symmetry breaking can account for the insulat-

ing phase that develops at the charge neutrality point of

near the magic angle. At strong and intermediate coupling, the opening of a gap between the lowest-energy valence and conduction bands proceeds preferentially through the breakdown of chiral symmetry. This effect is mainly driven by the extended component of the Coulomb interaction. When this is under very strong dielectric screening, however, we enter a regime where the on-site Coulomb repulsion becomes dominant, leading to the prevalence of time-reversal and parity symmetry breaking in a narrow weak-coupling window of the phase diagram.

Our study stresses the significance of the experimental conditions by which the Coulomb interaction is screened. In general, when the twisted bilayer is surrounded by nearby metallic gates, we find a competition between time-reversal and chiral symmetry breaking patterns as just mentioned. But in a more ideal case where the long-range tail of the Coulomb interaction is not significantly screened, we see the appearance of one more relevant pattern, which is characterized by breaking the time-reversal invariance but preserving parity. This gives rise to the superposition of two different effects, namely the opening of a gap at the Dirac cones and the splitting of the degeneracy of the low-energy bands at the $K$ points.

It would be interesting to investigate similar symmetry breaking effects at other filling levels of twisted bilayer graphene. This could clarify whether other strongly correlated phenomena may bear some connection with dynamical symmetry breaking, and whether this effect may be enhanced at fillings leading to a large density of states like that reached at the van Hove singularities in the low-energy bands.

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