Correlations in the elastic Landau level of a graphene/NbSe\(_2\) van der Waals heterostructure

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Electronic correlations stemming from nearly flat bands in van der Waals materials have demonstrated to be a powerful playground to engineer artificial quantum matter, including superconductors, correlated insulators and topological matter. This phenomenology has been experimentally observed in a variety of twisted van der Waals materials, such as graphene and dichalcogenide multilayers. Here we show that a hybrid graphene/dichalcogenide multilayer can yield a correlated state, emerging from an elastic pseudo Landau level. Our results build on top of recent experimental findings reporting that, when placed on top of a NbSe\(_2\) substrate, graphene sheets relax forming a periodic, long-range buckling pattern caused by the lattice mismatch. The low-energy physics can be accurately described by electrons in the presence of a pseudo-axial gauge field, leading to the formation of sublattice-polarized Landau levels. Moreover, we verify that the high density of states at the zeroth Landau level leads to the formation of a periodically modulated ferrimagnetic groundstate, which can be controlled by the application of external electric fields. Our results indicate that van der Waals heterostructures combining graphene and dichalcogenides are a versatile platform to explore emergent electronic states arising from correlated elastic Landau levels.

I. INTRODUCTION

One of the key features of graphene’s electronic structure is that low-energy electrons behave as massless Dirac fermions.\(^1,2\) Among the successful applications of this model, we can highlight the prediction of the so-called zeroth Landau level (ZLL) formed exactly at the Fermi energy,\(^3–5\) in contrast to the well-known behaviour for systems with parabolic low-energy dispersion.\(^6–8\) The high density of states resulting from the flat band dispersion leads to electronic instabilities at half-filling, e.g. the formation of canted antiferromagnetic ordering in the quantum Hall edge modes.\(^9–14\)

Interestingly, the emergence of Landau levels is not a unique consequence of orbital magnetic fields. They can also appear when the system is subjected to pseudo magnetic fields (PMF) and their corresponding pseudo-axial gauge fields, for example, due to the presence of strain,\(^15–20\) modulated interlayer hopping,\(^21\) or interlayer bias.\(^22\) In these artificial Landau levels, which also appear in twisted bilayer graphene systems displaying the so-called magic angle flat bands bands\(^21,23–25\) electronic instabilities are also present.\(^26–33\) The emergence of correlations in van der Waals superlattices has also been reported in a variety of Moiré graphene multilayers\(^34,35\) and Moiré dichalcogenide multilayers,\(^36,37\) suggesting that van der Waals systems combining both graphene and dichalcogenides can provide an additional new platform for correlated physics.

Here we put forward a minimal van der Waals graphene/dichalcogenide multilayer showing a correlated state, stemming from the emergence of localized modes associated to an elastic gauge field. Our results build on top of recent experimental reports regarding the formation of buckled graphene superlattices when the material is placed on top of a NbSe\(_2\) substrate.\(^38\) Indeed, the experimental data shows the formation of Landau subbands with sublattice polarization – a distinctive signature of PMF, suggesting a low energy description realizing a periodically-modulated pseudo-axial gauge field.\(^38,39\)

In this paper, we investigate the effects of electronic interactions in the pseudo Landau level of graphene/NbSe\(_2\), showing the emergence of localized correlated states. In particular, we show the emergence of a periodically-modulated ferrimagnetic groundstate, realizing an approximate magnetic honeycomb superlattice. We also consider the effects from the substrate, namely, charge doping and spin-orbit coupling (SOC). The first shows optimal magnetization for half-filling, while the second leads to a non-collinear ferrimagnetic ordering. Finally, we show that the presence of an external perpendicular electric field breaks the global sublattice symmetry as an effective mass in the superlattice scale, suppressing the magnetic ordering when this effective mass is comparable with the gap size.

The manuscript is organized as follows. Sec. II is devoted to introducing the non-interacting model. In Sec. III, we present key results regarding the magnetic ordering and the effects of the underlying NbSe\(_2\) substrate on both the band structure and the magnetic groundstate. Finally, in Sec. IV, we summarize our results.

II. THE SYSTEM

In this section, we consider a model for interacting electrons in graphene with a buckled superlattice, as described in Fig. 1 (a). The source code and data used to produce the figures in this work are available online.\(^40\)

Our starting point is the nearest neighbour tight-binding
The reduced Brillouin zone for the supercell is depicted in Fig. 1c. The interacting Hubbard term is solved at \( t/s \). We also need to rescale the Hubbard energy as \( U/t \). Unless explicitly written, all the results presented in this paper were obtained for a supercell with \( 25 \times 25 \) unit cells, corresponding to \( 25 \times 25 \) lattice. The colors represent the magnitude of the PMF, Eq. 5.

The low-energy description obtained by substituting Eq. 1 is the hopping energy and the one for free-standing graphene.

For the system under investigation, the corresponding PMF:

\[
B(\mathbf{r}) = B_{\text{eff}} \sum_{i=1}^{3} \cos(\mathbf{b}_i \cdot \mathbf{r}),
\]

with

\[
\mathbf{b}_1 = \frac{2\pi}{L_M} \left( \frac{1}{\sqrt{3}}, 1, 0 \right),
\]
\[
\mathbf{b}_2 = \frac{2\pi}{L_M} \left( \frac{2}{\sqrt{3}}, 0, 0 \right),
\]
\[
\mathbf{b}_3 = \frac{2\pi}{L_M} \left( \frac{1}{\sqrt{3}}, -1, 0 \right)
\]

can be implemented by the following gauge choice:

\[
A_x = 0, \quad A_y(\mathbf{r}) = B_{\text{eff}} \sum_{i=1}^{3} \cos(\mathbf{b}_i \cdot \mathbf{r} / \mathbf{b}_i \cdot \mathbf{e}_x),
\]

which is obtained by fixing the new hopping energies as:

\[
\Delta t_2 = \Delta t_3 = -\frac{eV_F}{2} A_y,
\]
\[
\Delta t_1 = \frac{eV_F}{2} A_y.
\]

The careful reader might notice that \( C_3 \) symmetry is slightly broken with such choice for hoppings. This is a similar problem to the breaking of translational symmetry when fixing gauge of a system with uniform magnetic field. Nevertheless, one might expect the results to be qualitatively the same and we will make it clear whenever this unwanted effect is visible in the plots.

Due to the rescaling of the system, we will measure the magnetic field in terms of the dimensionless parameter \( L_M/l_B \), where \( l_B = \sqrt{\hbar/eB_{\text{eff}}} \). The experiment shows \( L_M/l_B \sim 7 \). Unless explicitly written, all the calculations were performed for \( L_M/l_B = 5.5 \).

The emergence of a pseudo-axial gauge field can also be explicitly derived from the real space tight binding model Eq. 1 with hopping constants given by Eq. 2 without resorting to the low-energy description. For this purpose, we consider the real space valley flux \( \chi(\mathbf{r}) \) and define the valley Chern number of the system as its integral over the unit cell:

\[
C_V = C_K - C_{K'} = \int_{\text{u.c.}} \chi(\mathbf{r}) d^2\mathbf{r}.
\]

The real space valley flux in the tight binding model is equivalent to the analytically derived valley-dependent
magnetic field, and therefore will reflect the real space structure of the emergent magnetic field explicitly in the full tight binding model across the unit cell. The real-space valley flux can be computed within the Green’s function formalism as: \[ \chi(r) = \langle r | \int_{-\infty}^{0} d\omega \int_{BZ} \frac{d^2k}{(2\pi)^2} \frac{\epsilon_{\alpha\beta}}{2} G_V(\partial_{k_\alpha} G_V^{-1})(\partial_{k_\beta} G_V)|r\rangle. \] Here, \( \epsilon_{\alpha\beta} \) denotes the Levi-Civita tensor,

\[ G_V = [\omega - H(k) + i0^+]^{-1} P_V \]

the Green’s function associated to the Bloch Hamiltonian \( H(k) \), and \( P_V \) the valley polarization operator.22,43,44 As shown in Fig. 1 (d), it is clearly observed that certain regions of the system show a positive valley flux, whereas other negative flux. The positive valley flux is associated to the regions with compressive anisotropic strain, whereas the positive valley flux is associated with the tensile anisotropic strain. This is the very same phenomenology expected from the artificial magnetic field obtained with a low energy Dirac expansion, reinforcing the connection between the low energy model and the exact solution of the tight binding model.

We now study the electronic dispersion in the absence (Fig 2 (a)) and presence (Fig 2 (b)) of electronic interactions. In the non-interacting case, the system remains gapless even in the presence of modulated strain, but with a highly reduced Fermi velocity (Fig 2 (a)). Moreover, we can observe that the strain modulation does not create intervalley scattering by projecting the resulting band diagram onto the valley states by means of the valley polarization operator \( P_V \), see Fig 2 (a). Hence, valley is still a good quantum number. Interestingly, when interactions are turned on (Fig 2 (b)), a gap opens up in the electronic structure, stemming from an emergent magnetic state that slightly breaks sublattice symmetry of the electronic spectrum. In the following, we address in details the origin of this symmetry breaking.

III. STRAIN INDUCED MAGNETISM

A. Formation of periodically-modulated magnetization

To better understand the emergence of the correlated state, it is convenient to look at the spatial distribution of the low energy states in the absence of interactions (Fig. 3 (a)). As shown in Fig. 3 (a), the low energy bands are peaked in a slightly asymmetric honeycomb lattice. The spatial distribution of these states corresponds to the zones of the superlattice under the influence of a strong elastic gauge field. Hence, according to the previous low energy discussion, these regions would be associated to zero pseudo Landau levels. The localized states resulting from the buckling pattern indeed present non-zero magnetic order parameters when Hubbard interactions are considered. As expected, the magnetization (Fig. 3 (b)) correlates with the density of states of the non-interacting system (Fig. 3 (a)).

Figure 3 (b) shows the development of a periodically-modulated ferrimagnetic order parameter, which can also be interpreted as an antiferromagnetic signal with a noticeable sublattice imbalance due to the superposition of a small ferromagnetic signal, see Fig. 3 (c), in agreement with previous studies of a similar system.26 In fact, the global magnetization of the system is negligible, since the ferromagnetic ordering flips as the PMF changes in sign. Both of these facts are again a consequence of the global sublattice symmetry being broken only locally.

In order to properly define the dependence of the magnetization on the effective magnetic field, we must take into account that the latter changes in sign. Thus, we
Figure 3: (a) Local density of states for the non-interacting case and (b) magnetization along the $z$-direction revealing antiferromagnetic ordering for the interacting case with $U = 1.8t$ (note the small breaking of $C_3$ symmetry). A closer analysis reveals a ferrimagnetic periodic ordering, with the ferromagnetic component changing in sign with $B(r)$. (c) The dependence of both the modulated ferromagnetic ($\Xi$) and antiferromagnetic ($N$) order parameters on the PMF for a $10 \times 10$ supercell and $U = 2t$ indicating the emergence of an ordered groundstate for $L_M / \sqrt{3} l_B \approx 2$.

define the ferromagnetic magnetization as

$$\Xi = \left| \sum_i \text{sgn} [B(r_i)] (m_i \cdot e_z) \right|,$$

while the antiferromagnetic signal corresponds to the usual Néel order parameter

$$N = \left| \sum_i (m_i \cdot e_z) \sigma_i \right|,$$

where $m_i$ is the magnetic moment at position $i$ and $\sigma_i$ is the corresponding sublattice index $\pm 1$.

It is interesting to note that the magnetization profile shown in Fig. 3 (b) corresponds to an emerging honeycomb superlattice (actually, such emerging superlattice is already realized in the non-interacting LDOS profile in Fig. 3 (a)). Namely, we can distinguish two different regions with net magnetization $M_z > 0$ and $M_z < 0$ with majority of electrons located at $A$ and $B$ sublattices, respectively. Each of these regions can be understood as different Wannier orbitals of the emerging superlattice. Thus, all the phenomenology can be reduced to the analysis of such emerging honeycomb lattice. Also, the order parameter $\Xi$ can be understood as the Néel order parameter of such superstructure.

We show in Fig. 3 (c) the dependence of both magnetizations on the PMF, indicating a transition for $\eta := L_M / \sqrt{3} l_B \approx 2$, which means the transition occurs when the distance of the supercell Wannier orbitals, $L_M / \sqrt{3}$, is larger than the diameter of the cyclotron orbits, $2l_B$. As a matter of fact, previous results showed that the scale-independent parameter is actually the product between the PMF and the number of sites inside the supercell, a conclusion we verified for the system under consideration. Therefore, the magnetic groundstate is expected for PMFs within the experimental range.

**B. Effects of doping and spin-orbit coupling**

Since all the phenomena explored here are a consequence of interactions between a graphene sheet and a NbSe$_2$ substrate, it is important to consider the possible effects of the underlying material. The two most relevant effects induced by the proximity of two materials are: (i) electronic doping due to charge transfer and (ii) effects of spin-orbit coupling (SOC). The energy scales for both effects were already estimated by means of density functional theory (DFT).

Considering charge doping, one can check in Fig. 4 (c) a rapid decay in the magnetic ordering as one goes away from half-filling. DFT estimations, however, show that this is not the expected regime, but external bias can counterbalance this intrinsic doping effect to measure the ZLL bands.

We include spin-orbit coupling stemming from broken...
mirror symmetry with the substrate by adding a Rashba-like term to the graphene Hamiltonian:

\[ H_{SOC} = i \lambda R \sum_{i,j} (d_{ij} \times \sigma_{s,s'} c_{is}^{\dagger} c_{js'}) \cdot \hat{z}. \]  

(17)

The effects of SOC are quite significant. Spin-momentum coupling explicitly breaks the spin rotation symmetry, allowing in-plane contributions to the magnetization to appear, as shown in Fig. 4 (a). On the other hand, a net out-of-plane ferromagnetic ordering is present in the honeycomb superlattice, see Fig. 4 (b), indicating a resulting non-collinear ferrimagnetic ordering.

C. Breakdown of magnetic ordering with electric fields

The buckling pattern induces a non-homogeneous height variation of the graphene sheet with respect to the underlying substrate. Hence, the application of a perpendicular electric field should induce non-homogeneous energy shifts in real space. To account for this phenomenology, we consider the contribution from the following Hamiltonian:

\[ H_{elec} = \sum_{i=1}^{3} \mu(\mathbf{r}) c_{is}^{\dagger} c_{is} , \quad \mu(\mathbf{r}) = \mu_0 \sum_{i=1}^{3} \cos(\mathbf{b}_i \cdot \mathbf{r}) , \]  

(18)

where \( \mu_0 \) is proportional to the amplitude of the applied electric field.

The resulting band diagram in Fig. 5 (a) shows that, in the presence of such perpendicular electric field, a gap opens and global sublattice symmetry breaks. It is straightforward to understand this effect if we consider the emerging honeycomb superlattice: the electric field has the same effect as a sublattice mass in the superlattice. That said, one should expect that the magnetic ordering should be suppressed when the sublattice mass is larger than the antiferromagnetic gap. Indeed, that is exactly what we observe in Fig. 5 (b).

IV. CONCLUSION

We showed that the zeroth pseudo Landau level subband formed in buckled graphene superlattices on top of a NbSe\(_2\) substrate hosts a periodic magnetically ordered groundstate. This periodic pattern results in an emerging antiferromagnetic honeycomb superlattice. Moreover, we showed that in the non-interacting scenario, a perpendicular electric field opens up a gap, similarly to the interaction induced magnetic order. Interestingly, in the interacting case, the competition between the bias induced mass and the antiferromagnetic gap provides a route for electrically controlling the magnetic groundstate of the system. Our results show that strained graphene on top of NbSe\(_2\) provides a powerful platform to explore correlated physics in hybrid van der Waals materials, and to study the interplay between artificial gauge fields and interactions. Finally, it is worth noticing that the interplay of such magnetic state with the NbSe\(_2\) superconductivity, not addressed in the current manuscript, can lead to a versatile platform to explore superlattice Yu-Shiba-Rusinov physics, and ultimately Majorana states.

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