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Symmetric Kondo Lattice States in Doped Strained Twisted Bilayer Graphene

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We use the topological heavy fermion (THF) model [1] and its Kondo Lattice (KL) formulation [2] to study the possibility of a symmetric Kondo (SK) state in twisted bilayer graphene. Via a large-N approximation, we find a SK state in the KL model at fillings $\nu = 0, \pm 1, \pm 2$ where a KL model can be constructed [2]. In the SK state, all symmetries are preserved and the local moments are Kondo screened by the conduction electrons. At the mean-field level of the THF model at $\nu = 0, \pm 1, \pm 2, \pm 3$ we also find a similar symmetric state that is adiabatically connected to the symmetric Kondo state. We study the stability of the symmetric state by comparing its energy with the ordered (symmetry-breaking) states found in Ref. [1] and find the ordered states to have lower energy at $\nu = 0, \pm 1, \pm 2$. However, moving away from integer fillings by doping the light bands, our mean-field calculations find the energy difference between the ordered state and the symmetric state to be reduced, which suggests the loss of ordering and a tendency towards Kondo screening. In order to include many-body effects beyond the mean-field approximation, we also performed dynamical mean-field theory (DMFT) calculations on the THF model in the non-ordered phase. The spin susceptibility follows a Curie behavior at $\nu = 0, \pm 1, \pm 2$ down to ~ 2K where the onset of screening of the local moment becomes visible. This hints to very low Kondo temperatures at these fillings, in agreement with the outcome of our mean-field calculations. At non-integer filling $\nu = \pm 0.5, \pm 0.8, \pm 1.2$ DMFT shows deviations from a $1/T$-susceptibility at much higher temperatures, suggesting a more effective screening of local moments with doping. Finally, we study the effect of a C_{3z}-rotational-symmetry-breaking strain via mean-field approaches and find that a symmetric phase (that only breaks C_{3z} symmetry) can be stabilized at sufficiently large strain at $\nu = 0, \pm 1, \pm 2$. Our results suggest that a symmetric Kondo phase is strongly suppressed at integer fillings, but could be stabilized either at non-integer fillings or by applying strain.

Introduction— The experiments on magic-angle ($\theta = 1.05^\circ$) twisted bilayer graphene (MATBG) [3–5] have established the existence of a variety of interesting phases [6–27], including correlated insulating phases [28–38] and superconductivity [39–43]. Their discovery has been followed by considerable theoretical efforts [44–68] aimed at understanding their origin. An extended Hubbard model has been constructed to analyze the interacting physics [59, 69–81], however, due to the non-trivial topology of the flat bands [82–90], certain symmetries become non-local. Alternatively, an approach based on a momentum space model has been considered [91–99], in which correlated insulators [100–107], superconductivity [108–113], and other correlated quantum phases [114–118] have been identified and studied. Besides, various numerical calculations [119–126] have also been performed to investigate the correlated nature of the phenomena. However, the active phase diagram including the states at non-integer fillings is not well understood. The exact mapping between the MATBG and topological heavy-fermion model constructed in Ref. [1] could be used for developments in this direction. This mapping establishes a bridge between heavy-fermions [127–131] and moiré systems [1, 2, 132]. The presence of localized moments in MATBG is supported by recent entropy measurements which have found a Pomeranchuk-type transition [17, 18]. A large entropy observed at high temperatures, originates from weakly interacting local moments whose fluctuations are quenched at low temperatures [17, 18]. Since a similar behavior is observed in heavy fermion systems [127], where the fluctuating local moments are screened by conduction electrons (Kondo effect), this observation is suggestive of a Kondo state with screened local moments in MATBG.

In this work, we first use the KL model [2] to describe and study the symmetric Kondo (SK) state. The SK phase preserves all symmetries; the local moments are screened. We discuss the properties of the SK state and extend the study to the symmetric state in the THF model [1, 128]. To address the stability of the symmetric state, we perform both mean-field and DMFT calculations of the THF model. Our calculation indicates that, at integer fillings $\nu = 0, \pm 1, \pm 2$, ordered states are energetically favored with suppressed Kondo effect. Doping away from integer fillings tends to destroy the order and enhance the Kondo effect. We also study the effect of a C_{3z}-breaking strain. Our mean-field calculations show that

the ordering at $\nu = 0, \pm 1, \pm 2$ is suppressed by the strain effect and a symmetric state can be stabilized at a sufficiently large strain.

THF model and KL model— The THF model [1] contains two types of electrons: topological conduction c-electrons ($c_{\mathbf{k},a\eta s}$) and localized f-electrons ($f_{\mathbf{R},\alpha\eta s}$). The operator $c_{\mathbf{k},a\eta s}$ annihilates conduction c-electron with momentum $\mathbf{k}$, orbital $a \in \{1,2,3,4\}$, valley $\eta \in \{+,-\}$ and spin $s \in \{\uparrow,\downarrow\}$. We call the c-electrons with $a = 1,2$ ($a = 3,4$) as Γ_3 ($\Gamma_1 \oplus \Gamma_2$) c-electrons [1]. $f_{\mathbf{R},\alpha\eta s}$ is the annihilation operator of the f-electron at the moiré unit cell $\mathbf{R}$ with orbital $\alpha \in \{1,2\}$, valley η and spin s [1]. The Hamiltonian of the THF model [1, 133] is $\hat{H}_{THF} = \hat{H}_c + \hat{H}_{fc} + \hat{H}_U + \hat{H}_W + \hat{H}_V + \hat{H}_J$ with the values of parameters taken from Ref. [1]. $\hat{H}_c$ describes the kinetic term of conduction electrons and $\hat{H}_{fc}$ describes the hybridization between f-c electrons [1, 133]. The interactions include an on-site Hubbard interaction of f-electrons ($\hat{H}_U$), a repulsion between f- and c-electrons ($\hat{H}_W$), a Coulomb interaction between c-electrons ($\hat{H}_V$), and a ferromagnetic exchange coupling between f-and c-electrons ($\hat{H}_J$) [1, 133]. Based on the THF model [1], a KL model of MATBG has been constructed via the Schrieffer–Wolff transformation [2]. The KL model is described by $\hat{H}_{Kondo} = \hat{H}_c + \hat{H}_{cc} + \hat{H}_K + \hat{H}_J$ where $\hat{H}_{cc}, \hat{H}_K$ emerge from the SW transformation. $\hat{H}_{cc}$ is the one-body scattering term of Γ_3 c-electrons with the form of

$$\hat{H}_{cc} = \sum_{|\mathbf{k}|<\Lambda_c} \sum_{\substack{a,a'\in\{1,2\} \\ \eta,s}} e^{-|\mathbf{k}|^2\lambda^2} : c^\dagger_{\mathbf{k},a\eta s} c_{\mathbf{k},a'\eta s} : \left(\frac{-1}{D_{\nu_c,\nu_f}} + \frac{-1}{D_{\nu_c,\nu_f}} \right) \begin{bmatrix} \gamma^2/2 & \gamma v'_\star(\eta k_x - i k_y) \\ \gamma v'_\star(\eta k_x + i k_y) & \gamma^2/2 \end{bmatrix}_{a,a'} . \tag{1}$$

λ is the damping factor of the f-c hybridization in the THF model. $\gamma, v'_\star$ characterize f-c hybridization [1, 133]. D_{1,ν_c,ν_f} and D_{2,ν_c,ν_f} are two parameters given in Supplementary Materials (SM) [133]. $\hat{H}_K$ is the Kondo interaction between f- and Γ_3 c-electrons given in Sec II, SM [133]. We also note that, ground states at filling ν and $-\nu$ are connected by a charge-conjugation transformation [1]. This can be broken by other one-body terms which are expected to be small. In what follows, we only focus on $\nu \leq 0$.

Symmetric Kondo state– We perform a mean-field (large-N) study of the KL model [128], where the Kondo interaction is treated via a Hartree-Fock decoupling (see SM [133]) by introducing the hybridization fields

$$V_1^* = \sum_{\mathbf{R},|\mathbf{k}|<\Lambda_c} \sum_{\alpha\eta s} \frac{e^{i\mathbf{k}\cdot\mathbf{R} - |\mathbf{k}|^2\lambda^2/2}}{\sqrt{N_M} N_M} \langle \Psi | f^\dagger_{\mathbf{R},\alpha\eta s} c_{\mathbf{k},\alpha\eta s} | \Psi \rangle$$

$$V_2^* = \sum_{\mathbf{R},|\mathbf{k}|<\Lambda_c} \sum_{\alpha a\eta s} \frac{e^{i\mathbf{k}\cdot\mathbf{R} - |\mathbf{k}|^2\lambda^2/2}}{\sqrt{N_M} N_M} (\eta k_x \sigma_x + k_y \sigma_y)_{\alpha a} \langle \Psi | f^\dagger_{\mathbf{R},\alpha\eta s} c_{\mathbf{k},\alpha\eta s} | \Psi \rangle \tag{2}$$

with $|\Psi\rangle$ the mean-field ground state. This mean-field approach suppresses the RKKY interaction and essentially restores the hybridization term $\hat{H}_{fc}$ of the original periodic Anderson model but in a renormalized form [128, 134]. It becomes exact in the $N \to \infty$ limit (we have $N = 4$ which corresponds to the approximate flat $U(4)$ symmetry). By solving the mean-field equations at $\nu = 0, -1, -2$, we identify a SK state that preserves all the symmetries with $V_1 \neq 0, V_2 \neq 0$ [133].

We describe the properties of the SK state. In Fig. 1, we plot the band structure of the SK state and compare it with the non-interacting band structure of THF model. We find the f-c hybridization in the SK state to be enhanced. Consequently, the gap of the Γ_3 states at the Γ point [1] is increased from its non-interacting value 24.75meV at $\nu = 0$, to 168meV, 190meV, 213meV at $\nu = 0, -1, -2$ respectively. Furthermore, the flat bands in SK state are mostly formed by $\Gamma_1 \oplus \Gamma_2$ c-electrons with weights larger than 70%. The bandwidths of the flat bands at $\nu = -1, -2$ become 16meV, 53meV, which are (much) larger than the non-interacting bandwidth (7.4meV).

The flat bands in the SK state form the same representations as the flat bands in the non-interacting THF model [1]. The flat bands in SK state then belong to a fragile topology [1] at $\nu = -1, -2$. At $\nu = 0$, due to the additional particle-hole symmetry, flat bands have a stable topology and symmetry-protected nodes at Fermi energy [1, 84, 90] (see Sec IV, SM [133]). We also mention that the interplay between the Kondo effect and the topological bands has also been studied in various other systems [135–140].

Symmetric state in the THF model and the effect of doping— We next investigate the symmetric state in the THF model. We first focus on integer fillings $\nu = 0, -1, -2, -3$ and perform the mean-field calculations of the THF model (see Sec. V, SM [133]). We identify a symmetric state that preserves all the symmetries and is adiabatically connected to the SK state. To observe the stability of the symmetric state, we compare its energy (E_{sym}) with the energy (E_{order}) of the ordered (symmetry-breaking) ground states derived in Ref. [1]. The ordered ground states are a Kramers inter-valley-coherent (KIVC) state at $\nu = 0$, a KIVC+valley polarized (VP) state at $\nu = -1$, a KIVC state at $\nu = -2$ and a VP state at $\nu = -3$. However, we point out that at $\nu = -3$ translational-symmetry-broken states with lower energy exist [141]. In our calculations, $\Delta E = E_{sym} - E_{order} > 0$ at integer fillings indicate the ground states are ordered states instead of SK states at these fillings. However, introducing the Gutzwiller projector to our wave function may further reduce the energy of the symmetric state by including the many-body effect.

We next investigate the effects of doping. We stick to a narrow region $\nu \in [\nu_{int} - 0.5, \nu_{int} + 0.5]$ near each integer filling $\nu_{int} = 0, -1, -2, -3$ and compare the energies of the ordered states E_{order} and the symmetric states E_{sym} in the THF model. The ordered solution is generated by doping the ordered state at integer filling ν_{int} and performing self-consistent calculations (see Sec. V, SM [133]). Fig. 2 displays

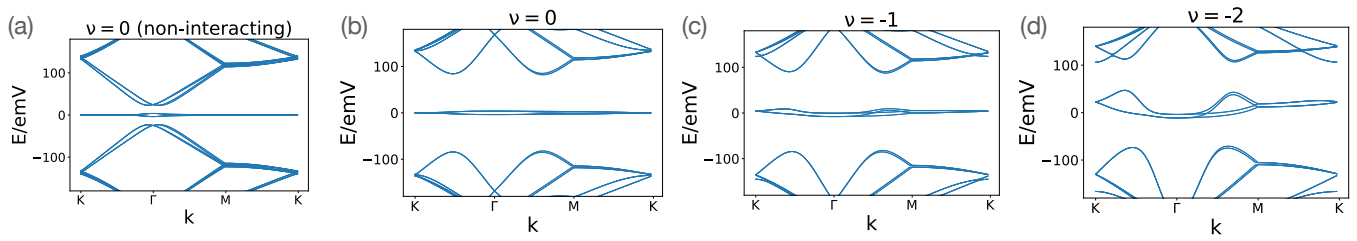

FIG. 1. (a) Band structure of the non-interacting THF model at $\nu = 0$. (b), (c), (d) Band structure of the SK phase at $\nu = 0, -1, -2$ respectively.

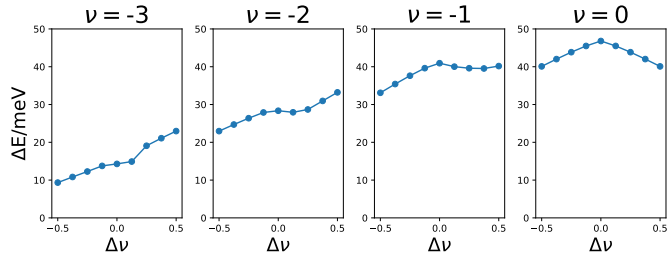

FIG. 2. Doping dependence of the ground state energy difference $\Delta E = E_{sym} - E_{order}$ near integer fillings $\nu = 0, -1, -2, -3$ in THF model. The ordered states we considered are KIVC ($\nu = 0$), KIVC+VP ($\nu = -1$), KIVC ($\nu = -2$) and VP ($\nu = -3$).

a plot of the difference of the ground state energies ΔE as a function of doping $\Delta\nu = \nu - \nu_{int}$. We observe that hole doping at $\nu = 0, -1, -2, -3$ and electron doping at $\nu = 0$ decreases the ΔE. Doping holes at $\nu = 0, -1, -2$ and doping electrons at $\nu = 0$ to the ordered states are equivalent to doping the light bands mostly formed by c-electrons [1, 133]. After doping, the conduction electrons stay close to the Fermi energy, and then enhance the tendency towards the Kondo effect and reduce ΔE.

However, doping electrons at $\nu = -1, -2$ is equivalent to doping heavy (flat) bands which mostly come from the f-electrons. Because of the flatness of the band, the nature of the ordered states will change with doping (see Sec. V, SM [133]). From our calculations near $\nu = -1, -2$, we find that ΔE will first decrease and then increase as we increase $\Delta\nu$. The change of order moments indicates the importance of the correlation effect which could be underestimated in the mean-field approach.

To further investigate the correlation effect, we perform DMFT calculations of the THF model in the non-ordered phase at both integer and non-integer fillings. DMFT finds a qualitative difference between the strong quasiparticle renormalization at integer filling and a Fermi liquid at non-integer fillings: this can be seen from the scattering rate $\Gamma_f = -\mathrm{Im}\Sigma_f(\omega = 0)$ which is shown as a function of the total filling ν at $T = 11.6$K in Fig.3(a). The largest scattering rates are found close to $\nu = 0.0$, -1.0 and -2.0, progressively decreasing as one moves away from the charge neutrality point. Correspondingly, the spectral weight at the Fermi level (black and grey solid circles) is suppressed at these fillings.

Fig.3(b) illustrates the temperature-dependent screening of the local magnetic moment on the f orbitals. We determine the screening temperature $T_\odot$ and the effective moment μ_{eff} by fitting the z-component of the local spin susceptibility to the expression $\mu_{\text{eff}}^2/3(T + 2T_\odot)$. A visualization of the deviation from Curie law can be obtained by plotting $T \cdot \chi^{\text{loc}}_{\text{spin}}(\omega = 0)$: a flat profile indicates Curie behavior and a well-defined effective local moment, while deviations signal the onset of screening and a crossover towards a Pauli-like behavior [142]. While at $\nu = 0.0$, -1.0 and -2.0 the $1/T$-like local spin susceptibility persists down to 1-2 K, the non-integer fillings deviate from Curie behavior at much higher temperatures($\sim 10K$). This deviation from Curie behavior at $\sim 10K$ also marks the the onset of the Kondo screening process. Our DMFT calculations suggest that the Kondo phase is strongly suppressed at integer fillings $\nu = 0, -1, -2$, increasing the propensity towards long-range order. By doping the system, the development of Kondo screening is observed, which suggests that doping could enhance the Kondo effect. This picture is consistent with our mean-field calculations.

Effects of strain— Since twisted bilayer graphene samples exhibit intrinsic strain [143] and the ordered states are disfavored by strain, we investigate the effect of strain in the THF model. We focus on $\nu = 0, -1, -2, -3$ and introduce a C_{3z}-symmetry breaking term [133, 144–146] to qualitatively characterizes the effect of strain :$\hat{H}_{strain} = \alpha \sum_{\mathbf{R},\eta s}(f^\dagger_{\mathbf{R},1\eta s} f_{\mathbf{R},2\eta s} + \text{h.c.})$ where α can reach ~ 10meV in real materials (Sec.V [133], SM). We compare the mean-field ground-state energies of the symmetric states (E^{strain}_{sym}) and the ordered states (E^{strain}_{order}) at non-zero strain. Due to the C_{3z} strain, the symmetric states here preserve all the symmetries except for the C_{3z}. In Fig. 4, we plot the energy difference $\Delta E^{strain} = E^{strain}_{sym} - E^{strain}_{order}$ as a function of the effective strain amplitude α. We observe ΔE at $\nu = 0, -1, -2$ vanishes at sufficiently large strain. A detailed analysis [133] of the wavefunction shows that the ordered state cannot be stabilized and converged to a symmetric solution at large strain. We thus conclude that a symmetric phase can be stabilized by sufficiently large strain at $\nu = 0, -1, -2$. As for $\nu = -3$, we mention again that other ordered states exist [141]. We leave a systematic analysis of $\nu = -3$ for future study. Finally, we comment that even at zero strain, a symmetric state that

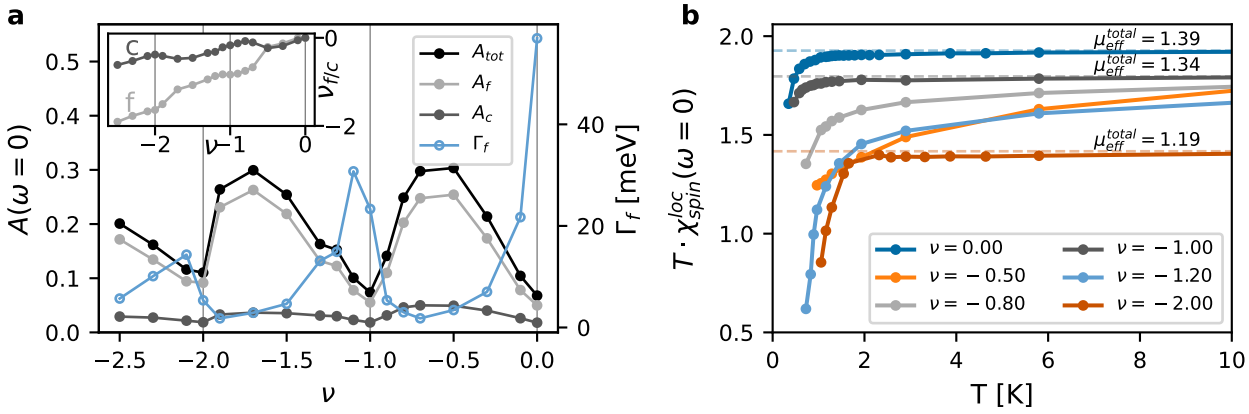

FIG. 3. DMFT solution of the THF model. (a) Doping ν dependent low-energy spectral function at the Fermi level ($A(\omega=0)$) for the full system A_{tot}, the c- (A_c) and the f-electrons (A_f) at 11.6 K. Also shown is the scattering rate Γ_f as extracted from the local f-electron self-energy. (b) Effective local moment $T\cdot\chi^{\rm loc}_{\rm spin}(\omega=0)$ as a function of temperature T for different doping levels ν. The full set of values of the screened local moments $\mu_{\rm eff}$ and screened temperature $T_\odot$ is provided in SM, Sec. VI [133].

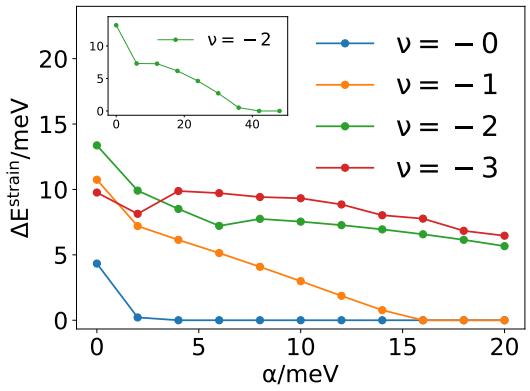

FIG. 4. Energy difference $\Delta E^{strain}=E^{strain}_{sym}-E^{strain}_{order}$ between the symmetric state that only breaks C_{3z} symmetry (E^{strain}_{sym}) and the ordered state (E^{strain}_{order}) as a function of α - a parameter characterizing the strain amplitude. (Inside) ΔE^{strain} at $\nu=-2$ over an extended parameter region 0meV $\leq\alpha\leq$ 45meV. We note that even at zero strain $\alpha=0$, a symmetric state that only breaks C_{3z} symmetry has lower energy than the fully symmetric state. We also note that there are small kinks due to the transition between two ordered phases at $\alpha\sim$ 3meV for $\nu=-1$, at $\alpha\sim$ 5meV for $\nu=-2$ and at $\alpha\sim$ 3meV for $\nu=-3$ (Sec. V, SM [133]).

breaks C_{3z} symmetry has lower energy than the fully symmetric state.

Discussion and summary— We have performed a systematic study of the SK state in the MATBG by analyzing its band structure, topology and stability. Our main result is that an ordered state, instead of a SK state, will be the ground state of the system at integer filling $\nu=0,-1,-2$. However, we find doping and C_{3z}-breaking strain can suppress the ordering and enhance the tendency towards the Kondo state. Most importantly, the possibility of the Kondo effect in the MATBG as we demonstrated leads to a natural explanation of entropy experiments [17, 18]. As has been established in the heavy-fermion systems [127], the presence of Kondo ground state leads to crossover behaviors from a fluctuating local-moment phase at high temperature to a Kondo phase with screened local moment at low temperature. The fluctuating local moments, which follow a Curie behavior, produce a large entropy accumulation at high temperature. However, at low temperature, the local moments are screened by the electrons via the Kondo effect [127] and the quenched fluctuations of the local moments lead to a small entropy. Remarkably, this is exactly what has been observed in the recent entropy experiments [17, 18], namely a large entropy at high temperature and a small entropy at low temperature.

Note added— After finishing this work, we learned that related, but not identical, results have also recently been obtained by the S. Das Sarma's [147], P. Coleman's [148], and Z. Song's groups [149]. We also mention that results from Z. Song's group are compatible with our DMFT results.

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