Impurity-induced triple point fermions in twisted bilayer graphene

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Triple point fermions are elusive electronic excitations that generalize Dirac and Weyl modes beyond the conventional high energy paradigm. Yet, finding real materials naturally hosting these excitations at the Fermi energy has remained challenging. Here we show that twisted bilayer graphene is a versatile platform to realize robust triple point fermions in two dimensions. In particular, we establish that the introduction of localized impurities lifts one of the two degenerate Dirac cones, yielding triple point fermions at charge neutrality. Furthermore, we show that the valley polarization is preserved for certain impurity locations in the moiré supercell for both weak and strong impurity potentials. We finally show that in the presence of interactions, a symmetry broken state with local magnetization can develop out of the triple point bands, which can be selectively controlled by electrostatic gating. Our results put forward twisted bilayer graphene as a simple solid-state platform to realize triple point fermions at charge neutrality, and demonstrate the non-trivial role of impurities in moire systems.

Topological semimetals have attracted a lot of attention in the past years, as they provide solid-state platforms to realize analogs of relativistic particles \cite{1, 2}, namely Dirac and Weyl fermions \cite{3}, whose spinorial form stems from Lorentz invariance. However, space group symmetries in materials provide an even more versatile playground, as they impose only a subset of the symmetries inflicted by Lorentz invariance, enabling novel types of effective particles to emerge beyond the conventional high energy paradigm \cite{4–19}. Among these possibilities, triple point fermions \cite{4, 5, 20–28} are exotic excitations displaying unusual magneto-transport phenomena including large negative magneto-resistance and helical anomaly \cite{20, 31}, in contrast to the chiral anomaly observed in Weyl semimetals \cite{3}. From the material science point of view, recent proposals suggest the presence of triple point fermions away from the Fermi energy in Heusler compounds \cite{20, 32}. Experimentally, ARPES measurements have observed triple point fermions \cite{13, 14, 15}, but only weak signatures of the expected exotic transport phenomena could be observed given the distance of the triple points to the Fermi level \cite{22, 30}. Thus, materials displaying robust triple point fermions at the Fermi energy have remained elusive, frustrating the experimental exploration of their associated exotic properties.

Graphene is known for being an extremely clean platform to explore Dirac fermion phenomena, with two spin degenerate Dirac cones at each $K$ point \cite{33}. Engineering triple point fermions out of graphene by lifting the degeneracy of the Dirac points would require eliminating key symmetries, which usually leads to a shift of the cones or to a gapped spectrum. Introducing an additional level of complexity, graphene multilayers have the potential to enlarge the degeneracy of the Dirac cones, providing new routes for degeneracy lifting. Among them, twisted bilayer graphene (TBG) (Fig. 1(a)) is a specially promising candidate for displaying a band structure with four-fold degenerate Dirac cones in the reduced Brillouin zone (Fig. 1(b,c)) \cite{34, 37}, providing a potential direction for the engineering of triple points by the controlled reduction of symmetries.

![FIG. 1.](image-url) (a) Perspective view of twisted bilayer graphene, highlighting the electronic density concentrated in the AA regions (yellow blobs), the AB/BA regions (orange/blue triangles), and the moire length $L_M$. (b) Brillouin zones for the first (pink) and second (green) layer, and the emergent moire Brillouin zone (black), with the respective $K$ and $K'$ points. (c) Sketch of the effects of a weak impurity and a vacancy on the degenerate low energy Dirac cones, with the generation of a triple point at the Fermi level.
In this Letter, we show that impurities in TBG create robust triple point fermions. In particular, we show that the interplay of the Dirac point degeneracy and the local nature of the impurity gives rise to mass generation in one of the Dirac cones, creating a triple point at charge neutrality independently of the strength and location of the impurity (Fig. 1(c)). We show that the valley polarization of the triple point can be controlled by the location of the impurities in the moire supercell, and we address the interplay of interaction-driven symmetry broken states in this graphene-based triple point system.

We focus on TBG superlattices with long moire wavelength, i.e. small twisting angles $\alpha = 1^\circ - 2^\circ$ (see Fig. 1 (a)), and for the sake of simplicity, we omit the spin degree of freedom. In this regime, the low energy model consists of two sets of honeycomb-like bands with strongly renormalized Fermi velocity, which vanishes at the magic angle $\alpha \approx 1^\circ$ [37, 38]. We model TBG in the presence of impurities by the real space Hamiltonian of the form:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{W},$$

where $\mathcal{H}_0$ encodes the pristine TBG tight binding Hamiltonian [39]:

$$\mathcal{H}_0 = t \sum_{\langle i,j \rangle} c_i^\dagger c_j + \sum_{i,j} t_{\perp}(\mathbf{r}_i, \mathbf{r}_j)c_i^\dagger c_j,$$

and $\mathcal{W}$ describes a local impurity at site $n$

$$\mathcal{W} = wc_n^\dagger c_n$$

where $c_i^\dagger (c_i)$ is the fermionic creation (annihilation) operator at site $i$, $w$ is the impurity potential strength, $t$ is the nearest neighbor hopping, $\langle i,j \rangle$ indicates the sum over first neighbors, $t_{\perp}(\mathbf{r}_i, \mathbf{r}_j)$ is the distance-dependent interlayer coupling taking a maximum value $t_{\perp}$ for perfect stacking [40]. As a reference, the values of the parameters in graphene are $t \approx 3$ eV and $t_{\perp} \approx 300$ meV [41]. The previous Hamiltonian is defined in a moire unit cell with $N = 4(3m_0^2+3m_0+1)$ sites, with $m_0$ an integer, and the magic angle regime is reached for $t_{\perp}/(tm_0) \approx 0.025$.

In the absence of impurities, $w = 0$, the low energy spectra of the previous Hamiltonian consists of two Dirac cones at $K$ and other two at $K'$ [33, 34, 42]. This degeneracy can be understood by the folding of the cone at $K_1$ from layer 1 and $K_2'$ from layer 2 at the same $K$ point in the moire Brillouin zone, as can be seen from Fig. 1 (b). Due to the approximate valley symmetry of the low energy graphene Hamiltonian, the states associated with each decoupled layer $\ell$ in TBG can be labeled by their valley number $\nu_\ell$. In a real space tight-binding formalism, such valley flavor can be computed using the valley operator: $\nu_\ell = \frac{1}{3\sqrt{2}} \sum_{\langle i,j \rangle} \psi_\ell h_{ij} \sigma_z^{ij} c_i^\dagger c_j$, where $\langle i,j \rangle$ denotes second neighbor sites, $h_{ij} = \pm 1$ for clockwise or anticlockwise hopping, and $\sigma_z^{ij}$ is a Pauli matrix associated with the sublattice degree of freedom [43, 44].

The interlayer hopping couples opposite valleys between the two layers, giving rise to a new quantum number $\nu = \nu_1 - \nu_2$, which is conserved in the absence of impurities. The addition of impurities generally introduces inter-valley scattering, therefore it is interesting to track the valley polarization of the states.

When the local impurity potential is turned on, $w \neq 0$, the four-fold degeneracy of the states at $K$ and $K'$ is lifted, giving rise to triple points at the Fermi level, as shown in Fig. 2 (a), independently of the position of the impurity in the moire pattern. The location of the impurity controls the splitting $\Lambda$ between the triple point and the higher lying state $\Psi_\Lambda$, at the $K$ point, as a function of the impurity position. Here we took $m_0 = 11$, $t_{\perp} = 0.3t$ and $w = 0.5t$.

![FIG. 2. (a) Band structure of twisted bilayer graphene with $\alpha = 1.5^\circ$ in the presence of a weak impurity in the AB region, showing the valley polarization $\langle \nu \rangle_\psi$ of each state (color gradient) and the emergence of a gap of magnitude $\Lambda$. (b) Map of the splitting $\Lambda$ as a function of the impurity position. (c) Map of the valley polarization of the higher lying state, $\Psi_\Lambda$, at the K point, as a function of the impurity position.](image-url)

The robustness of the triple point can be understood from the consideration of the band degeneracy of pris-
tine TBG. The band structure is displayed in Fig. 3 (a), with two double degenerate bands crossing the Fermi level in the $\Gamma - K - M$ direction [46]. We can label the eigenstates associated with the branches $E_{1k}$ as $\Psi_{1k}$ and $\Psi_{1k}$, and the other two associated with $E_{2k} = -E_{1k}$ as $\Psi_{2k}$ and $\Psi_{2k}$. These wave functions have weight in all microscopic degrees of freedom, layer and sublattice, such that these are expected to have a finite amplitude at a generic impurity site. Focusing on the first degenerate set, an impurity introduces a coupling between the eigenstates of pristine TBG such that the Hamiltonian in the eigenbasis $(\Psi_{1k}, \Psi_{1k})$ can be written as $H_{1}(k) = \left( \begin{array}{cc} E_{1k} + v_{1} & \sqrt{v_{1}v_{2}} \\ \sqrt{v_{1}v_{2}} & E_{2k} + v_{1} \end{array} \right)$, where $v_{1}$ and $v_{2}$ stand for the coupling of the impurity to the respective state, which are proportional to its amplitude squared at the site of the impurity, $v_{i} = \sum_{n} \left| \langle \Psi_{i} | \Psi_{n} \rangle \right|^{2}$. This eigenproblem has solution $\phi_{ik}$ with $E_{1k}$ and $\phi_{ik}$ with $E_{1k} + v_{1} + v_{2}$. Note that $\phi_{ik}$ remains at its original energy, indicating that it has zero amplitude at the impurity site and is therefore blind to its presence, while $\phi_{ik}$ couples to the impurity and is shifted in energy. An analogous construction can be made for states $\Psi_{2k}$ and $\Psi_{2k}$, such that in the appropriate combination, one of the states decouples from the impurity site, such that $\phi_{ik}$ and $\phi_{ik}$ give rise to two bands which do not change in presence of the impurity.

We now focus on the $K$ point, at which all low lying bands become degenerate. At this point, we can choose to write the eigenstates in a basis which is valley polarized. From the numerical analysis, we infer that this basis can, in fact, be written as $\Psi = \left( \phi_{1k}, \phi_{1k}, \phi_{2k}, \phi_{2k} \right)$, in terms of the eigenstates discussed in the previous paragraph. In this basis the valley operator yields $\mathcal{V} = \text{diag}(+1, +1, -1, -1)$, and the effective Hamiltonian around the $K$ point is of the form:

$$H^{\text{eff}}(K + k) = \begin{pmatrix} \Lambda_{1} & \bar{k} & 0 & \sqrt{\Lambda_{1}\Lambda_{2}} \\ \bar{k}^{*} & 0 & 0 & 0 \\ 0 & 0 & 0 & \bar{k}^{*} \\ \sqrt{\Lambda_{1}\Lambda_{2}} & 0 & \bar{k} & \Lambda_{2} \end{pmatrix} \tag{4}$$

where $\bar{k} = \bar{v}_{F}(k_{x} + ik_{y})$, $\bar{v}_{F}$ is the renormalized Fermi velocity, and $\Lambda_{i} = v_{i} + v_{i}$. The effective Hamiltonian at $K'$ follows from time reversal symmetry. In the absence of impurities, $\Lambda_{1,2} = 0$, the doubly degenerate Dirac dispersion is found. In the presence of an impurity, $\Lambda_{1,2} \neq 0$, the effective Hamiltonian has eigenvalues $\pm |\bar{k}|$ and $\frac{1}{2}(\Lambda \pm \sqrt{4|\bar{k}|^{2} + \Lambda^{2}})$, where $\Lambda = \Lambda_{1} + \Lambda_{2}$. Note that at the $K$ point ($\bar{k} = 0$) the eigenvalues are $\{0, 0, 0, \Lambda\}$, making the triple point explicit and associating the splitting to the magnitude of the impurity potential as $\Lambda \sim w$. In this generic scenario, states with different valley number are mixed. Note that, in case $\Lambda_{2} = 0$ and $\Lambda_{1} = 0$, a triple point emerges with well defined valley number, since $\left[ \mathcal{V}, H^{\text{eff}} \right] = 0$, as shown in Fig. 2 (a), which happens for impurities in the AB/BA regions, as mapped in Fig. 3 (c). This valley polarization for impurities in the AB/BA regions can be inferred from the properties of the Wannier wave functions in real space as shown for example in Ref. [55].

We now move on to consider the case of vacancy effects [47–55], namely $w \to \infty$. This limit is especially attractive because it physically represents adsorbed hydrogen atoms, which have been proven to be easily manipulated with extreme precision by scanning tunneling microscopy [57]. A vacancy in monolayer graphene is known to give rise to a zero mode at charge neutrality, according to Lieb’s theorem [55]. In TBG, this zero mode will, however, coexist in energy with the nearly flat honeycomb-like bands of Fig. 3 (a,b), and therefore it is expected to heavily hybridize with them. Interestingly, as shown in Fig. 3 (c), the hybridization of the vacancy mode with the honeycomb-like bands lifts one
of the Dirac cones and generates a flat band. Moreover, even though vacancies are expected to create strong inter-valley scattering, we find that the remaining Dirac cones are perfectly valley polarized when the vacancy is located at the AB/BA regions, see Fig. 3 (c,d) and (e,f), similarly to the weak impurity scenario above. The weak and strong potential limits can be actually continuously connected by ramping up the parameter \( w \). In this process, it is observed that the \( K \) point always displays a triple point, two of the bands remain rigid, and the quadratic band pinned to the Fermi level evolves smoothly towards a flat band. As the potential is increased, part of the electronic density drifts from the AA regions to the location of the impurity. This phenomenology is observed to be independent on the density of vacancies per unit cell, as shown in the calculation for a 2x2 supercell in Fig. 3 (e,f) \[59\].

It is important to note that this discussion relied on a single particle picture. However, the large density of states associated with these nearly flat bands suggests that, at low temperatures, a symmetry broken state develops due to interactions \[59, 60-63\]. To account for the effect of electronic interactions, we make explicit the spin degree of freedom in Eq. 1 and introduce an interaction term of the form \( H_I = U \sum_i n_{i\uparrow} n_{i\downarrow} \), where \( n_{i\sigma} \) counts the number of electrons with spin \( \sigma = \{\uparrow, \downarrow\} \) at site \( i \) \[45, 60, 61, 65\]. We use a mean field ansatz of the form \( H_F = V \sum_{\delta} n_i \delta n_j \), with the expectation values determined self-consistently, allowing for a local ground state magnetization \( m^z_i = \langle n_{i\uparrow} \rangle - \langle n_{i\downarrow} \rangle \).

For pristine TBG at half filling, Fig. 4 (a,b) show that interactions drive the system into an insulating state with antiferromagnetic order in the AA regions \[45, 60, 66\]. A more interesting scenario takes place in the presence of a vacancy in an AB region of TBG, in which case electronic interactions create a localized magnetic moment \[47-54\]. As shown in Fig. 4 (c,d), the impurity state is not detrimental to the opening of a gap and the associated antiferromagnetic ordering of the AA regions, even though the magnetization associated with the impurity states is one order of magnitude larger than the staggered magnetization in the AA regions (see inset). As a result of the weak antiferromagnetism in the AA regions, doping the system quenches the antiferromagnetic order, while the magnetization around the vacancy survives. This can be clearly seen in Fig. 4 (e) and (f), where we consider a doping of four extra electrons per unit cell, which fills the low energy bands up to their edge. In fact, this phenomenology holds up to chemical potentials of the order of the exchange splitting \( \sim 30 \text{ meV} \) \[49, 57\], much bigger than the \( \sim 8 \text{ meV} \) bandwidth of the nearly flat bands \[35, 36, 38, 42\]. As a result, at low temperatures, these localized magnetic moments may coexist with other phases found in the bilayer such as superconducting \[57\], strange metal phases \[68\] or anomalous Hall states \[69\].

To summarize, we have established that impurities in small-angle twisted bilayer graphene give rise to robust triple point fermions at the charge neutrality point, independently of the impurity potential and its location. We have shown that the triple point modes can be valley polarized for defects located in the AB and BA regions, providing a route to engineer triple-point fermions with an additional quantized degree of freedom. In the presence of interactions, the triple points can be lifted by the development of magnetic order, introducing the possibility of engineering correlated states of triple point fermions in twisted bilayer graphene. Our results put forward a new mechanism to generate triple-points in graphene systems, providing a starting point to study their intrinsic properties and interplay with additional emergent states in twisted bilayers.

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