Emergent symmetry and near-field chirality in twisted bilayer graphene

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The real-space chirality of twisted bilayer graphene close to the magic angle is discussed and a sign-change due to an emergent $C_6$-symmetry is observed. This collapse occurs not only for doping levels related to the first valence and conduction band, but extends up to 75meV. Our observation thus offers a new definition of the magic angle based on a macroscopic observable which is accessible in typical transport experiments. We further show that the same observable gives rise to an unprecedented large local chirality for the plasmonic near-field if the twist angle is slightly away from the magic angle. Twisted bilayer graphene or other twisted two-dimensional heterostructures might thus provide a novel platform to catalyze the reaction of new chiral molecules while conserving time-reversal symmetry.

Introduction. Magic-angle graphene, i.e., twisted bilayer graphene\textsuperscript{1–8} (TBG) at an angle of $\theta \sim 1.08^\circ$, has attracted tremendous attention since the discovery of correlated insulator states\textsuperscript{9,10} as well as superconductivity.\textsuperscript{11–13} Moreover, it represents the first material where Coulomb interactions can drive a system into a topologically non-trivial state leading to a valley and spin-polarised Chern band displaying anomalous ferromagnetism.\textsuperscript{14–16} This makes TBG and also related systems such as ABC-trilayer graphene on a misaligned BN-substrate\textsuperscript{17,18} an ideal platform to study the interplay between correlations and topology.

Additionally, plasmonics in TBG has received considerable interest\textsuperscript{19–21} and excitonic collective modes around the charge neutrality point have been predicted\textsuperscript{22} and observed.\textsuperscript{23} Furthermore, for small band-widths plasmons are expected to be long-lived\textsuperscript{24} and for twist angles of less than half a degree, the possibility of a photonic-crystal for these collective excitations opens up.\textsuperscript{25} Scanned probe optical techniques can also be used to determine the local twist angle and domain structure.\textsuperscript{26}

TBG displays an inherent real-space chirality depending on whether the twist angle is rotated clockwise or anti-clockwise. This chiral structure has direct experimental consequences and led to the observation of an intrinsic optical dichroism in TBG in the absence of symmetry-breaking fields.\textsuperscript{27–29} The system is thus capable to sufficiently rotate incidence light in between the two misaligned layers even though they are only 3.4Å apart. For this effect, the two layers need to be strongly coupled and, therefore, it is largest for frequencies that match the energy of the van Hove singularity, i.e., where the bands of the upper and lower layer cross and interact. In twisted boron nitride or transition metal dichalcogenides, the optical dichroism should further depend on the stacking type.\textsuperscript{30}

In Refs.\textsuperscript{31,32}, we have demonstrated that the chirality also has consequences for dc transport. The adiabatic application of an in-plane magnetic field, e.g., provokes a current flowing in the parallel or antiparallel direction depending on whether the system has positive or negative chirality. This was coined the longitudinal Hall effect because the chirality changes with the sign of the carriers just as in the ordinary Hall effect. Driving TBG by a

FIG. 1. Upper panels: Density of States (left) and chiral Drude weight in units $D_{xy}$ as function of the twist angle $\theta$ and Fermi energy $E_F$. For $D_{xy}$, we observe a sign change indicated by the black line. Lower panels: The chiral Drude weight in units of $t(c/h)^2$ for large twist angles with $i = 10, 15, 20$ (left) and for small twist angles with $i = 26, 28, 30, 32, 34$ (right) as function of the Fermi energy $E_F$. Twist angles are parametrised by $\cos \theta_i = \frac{3\pi^2 + 3\pi + 1/2}{3\pi^2 + 3\pi + 1}$ and $t = 2.78$eV denotes the in-plane hopping parameter.
source-drain bias can further lead to an emergent magnetic texture.\textsuperscript{33}

From the above introduction, one would conclude that for a given carrier type the chirality is uniquely linked to the sign of the twist angle, i.e., to the rotational sense of the underlying crystallographic structure. Nevertheless, in this Letter we will report an abrupt sign change at the magic angle for low enough temperature and this change in chirality further extends over a wide energy range, i.e., it is not limited to the lowest conduction/valence band. We also point out that the real-space chirality gives rise to chiral plasmonic near-fields that are strongly enhanced compared to circularly polarized light.

**Chiral Drude weight.** We seek an observable that characterizes the chirality of the electronic subsystem system of twisted bilayer graphene. As we will show, this chirality does not necessarily have to coincide with the chirality of the underlying atomic system.

The chirality $D_{xy}$ shall be given by the generalised density of states

$$D_{xy}(\mu) = \frac{1}{2A} \sum_{k,n} e_z \cdot (\mathbf{j}_{k,n} \times \mathbf{j}_{k,n}^*) \delta(\epsilon_{k,n} - \mu),$$ \hspace{1cm} (1)

where $\mathbf{j}_{k,n} = (k, n) \mathbf{j}(k, n)$ and $\epsilon_{k,n}$ and $(k, n)$ denote the eigenvalues and eigenvectors, respectively, with $k$ inside the first Brillouin zone. $A$ labels the area of the sample and $j_{\ell}^i$ is the current operator of layer $\ell = 1, 2$ in the direction $i = x, y$. The above definition is valid for the continuum model\textsuperscript{34} and implies that $D_{xy}$ is an odd function with respect to the twist angle $\theta$. For the particle-hole symmetric continuum model, one further has the exact symmetry $D_{xy}(\mu) = -D_{xy}(-\mu)$.

Eq. (1) is the low frequency limit of the current-current correlation function $\chi_{j_{\ell},j_{\ell}^*}(\omega) = -\frac{i}{\pi} \int_{-\infty}^{+\infty} dt e^{i\omega t} \langle [j_{\ell}^i(t), j_{\ell}^i(0)] \rangle$, i.e., $D_{xy} = \lim_{\omega \rightarrow 0} \chi_{j_{\ell},j_{\ell}^*}(\omega)$, as was discussed in Ref. 32. $D_{xy}$ shall, therefore, also be denoted as chiral or Hall Drude weight. Similarly, we can discuss the total Drude weight defined by $D_{\parallel} = \lim_{\omega \rightarrow 0} \chi_{j_{\parallel},j_{\parallel}^*}(\omega)$ with the total current $j_{\parallel} = j_{\parallel}^x + j_{\parallel}^y$.

Let us recall that the Hall Drude weight gives rise to the longitudinal Hall effect: \textsuperscript{31}

$$j = -aD_{xy} B,$$ \hspace{1cm} (2)

where $a \approx 3.4\text{Å}$ is the distance between the two layers and $B$ is the in-plane magnetic field causing the longitudinal current $j$. This is not an equilibrium property, but the response to the adiabatic introduction of the field, thus accessible to slow driving transport experiments in clean samples.

The results for $D_{xy}$ as function of the Fermi energy are obtained from the continuum model of TBG\textsuperscript{1,7} and shown in Fig. 1. For large angles, there is a sign change at the neutrality point as the carrier type changes from holes to electrons reminiscent of the ordinary Hall effect. But for the twist angle with $i = 30$ where $\cos \theta_i = \frac{3\ell^2 + 3\ell + 1/2}{3\ell^2 + 3\ell + 1}$, i.e., $\theta_{30} = 1.08^\circ$, we observe an abrupt change in chirality for Fermi energies up to $E_F \sim \pm 75$meV corresponding to the second valence and conduction band. This energy scale also coincides with the second van Hove singularity branch of the Density of States (DOS) at twist angle $\theta_{30}$ and a fractal structure develops in both the DOS and $D_{xy}$ for lower twist angles. For $D_{xy}$, there are further sign changes inside the dome which define higher-order magic angles. In contrary to the first magic angle, these do not coincide with those obtained in Ref. 7.

The spectrum of two different enantiomers is identical, but the change of chirality can be related to an emergent $C_6$-symmetry that is seen in the band structure and discussed in detail in the Supplemental Material.\textsuperscript{35} Usually, parity is broken in one valley and the bands only display a $C_3$-symmetry. Moreover, valence and conduction bands show dual $C_4$-symmetries which suggests a sign change at the neutrality point. Also the two valleys are related via dual $C_3$-symmetries, however, the valleys do not cancel, but add up to yield a finite $D_{xy}$.

With an emergent $C_6$-symmetry, the chiral Drude weight vanishes for Fermi energies within the first VB (CB). Interestingly, there is also an almost perfect cancellation of $D_{xy}$ coming from the second VB (CB) by the one from the third VB (CB).\textsuperscript{35} Furthermore, the interplay between the second and third band gives rise to an almost quantized plateau for commensurate angles labeled by $i$.

Let us note that the chirality has been calculated for the CM with equal interlayer hopping in the AA- and AB-region at low temperature. At room temperature, the change in chirality around the magic angle is lost. Gradually reducing the hopping in the AA-region leads to a reduction of $D_{xy}$ until it vanishes for all twist angles, i.e., the $C_6$-symmetry has been recovered for $t_{AA} = 0$.\textsuperscript{41}

**Near-field properties of twisted bilayer graphene.** Chiral plasmonics in twisted bilayer graphene has first been introduced and discussed in the non-retarded limit.\textsuperscript{31,32} This is usually enough since the transversal (p-polarised) current sources are suppressed by the fine-structure constant and can thus be neglected in comparison to the longitudinal (p-polarised) current sources. However, there are quantities which are zero in the non-retarded regime and these shall be discussed in this work within the full retarded response theory.

The plasmonic field in a general bilayer is generated by the in-plane currents $j = j_\parallel [\epsilon_\parallel + j_\parallel^x e_\parallel]$ with $\parallel = 1, 2$ that carry the momentum $\mathbf{q} = q[\epsilon_\parallel + j_\parallel^x e_\parallel]$ in the non-retarded limit. TBG displays chirality without breaking time-reversal symmetry and we have for the charged plasmon\textsuperscript{31,32} $j_\parallel \equiv j_\parallel^x + j_\parallel^y$ and $j_\perp \equiv j_\perp^x = -j_\perp^y$. This polarization of currents is for symmetric environments and we expect it to also roughly hold in asymmetric setups such as the one for screened plasmons discussed below.

Let us now explicitly consider two different dielectrics $\epsilon_i, \mu_i$ with $i = 1, 2$ in the two half-planes $|z| > a/2$. We are mainly interested in the near-field chirality that couples the longitudinal and transverse field component. For real electromagnetic fields ($\mathbf{E}, \mathbf{B}$) in a dielectric medium
\( (\epsilon, \mu) \), this is given by\(^{42} \)

\[
C = \frac{\epsilon \epsilon_0}{2} \mathbf{E} \cdot (\nabla \times \mathbf{E}) + \frac{1}{2 \mu \mu_0} \mathbf{B} \cdot (\nabla \times \mathbf{B}) ,
\]

(3)

which is related to the flux of chirality via the continuity equation \( \partial_t C + \mathbf{v} \cdot \mathbf{J} = 0 \). For TBG, we have

\[
C_i = -\frac{\mu_\mu_0 a q \text{e}^{2j_\parallel |\mathbf{E}|}}{2} ,
\]

(4)

\[
\mathbf{F}_i = -\frac{\mu_\mu_0 \omega q}{2} \text{e}^{-2q' |z|} \times \left[ 2 j_\parallel q' \text{e}^{2q' |z|} + \text{sgn}(z) j_\parallel' \left( \frac{q'}{k_i^2} \right)^2 \left( 1 + j_\perp \right) \right] ,
\]

(5)

where we defined \( j_\perp = 1 + \frac{j_\parallel^2 k_\parallel^2}{(q')^2} \) and \( k_i = \omega/c_i \) with \( c_i = c/\sqrt{\epsilon_i \mu_i} \) the speed of light of the dielectric medium. Other quantities such as the helicity and ellipticity can be obtained from the above expressions, see SM.\(^{35} \) Also notice that the chirality flux contains a non-trivial transverse component which could be chosen arbitrarily without violating the continuity equation. The local definition of the SM thus goes beyond the transport properties.\(^{43} \)

**Local chirality.** We can now relate the longitudinal and transverse current, given for the charged plasmon by \( j_\parallel = -2 \frac{D_{xy}}{\omega c} j_\parallel \) where \( D_T = 2(D_0 + D_1) \) defines the total Drude weight, introduced in Refs.\(^{31,32} \). For unscreened plasmons, we then have \( \omega^2 = q D_T/(\epsilon_0 (c_1 + c_2)) \). With \( 2 j_\parallel = -D_T A_\parallel \) and \( E = i \omega A_\parallel \), we obtain the final expression:

\[
C_i = -\frac{\epsilon_1 + \epsilon_2}{4 \epsilon_i} \frac{\alpha_D a D_{xy}}{D_T} k_i E^2 \text{e}^{-2q' |z|} .
\]

(6)

For screened (acoustic) plasmons, we have \( \omega^2 = \alpha_D a D_T/(\epsilon_0 c_2) \) where \( d \) and \( c_2 \) is the distance and the dielectric constant of the bilayer to the metallic plate, respectively.\(^{45} \) Using Eq. (35) with \( 2 j_\parallel = -D_T/A_\parallel \) and \( E = i \omega A_\parallel \), we obtain the final expression:

\[
\mathcal{C}_i = \frac{2 \epsilon_2}{4 \epsilon_i c_{i} c_{i} w d} |z| q_i \text{e}^{-2q' |z|} .
\]

(7)

**Comparison.** Let us contrast the near-field results with the chirality obtained for left (right) CPL with \( E_\parallel = E_0(1, \pm i, 0, 0) \text{e}^{i k_i z} \) which yields \( \mathcal{C}_i^0 = \pm \epsilon_i \epsilon_0 k_i E_0^2 \) and \( \mathcal{F}_i = c_i C_0 \epsilon_2 z \). For unscreened plasmons, we have for \( |z| q_i \ll 1 \)

\[
|\mathcal{C}_i/\mathcal{C}_i^0| = c_i a k_i \chi F^2 ,
\]

(8)

with the reduced chirality \( \chi = D_{xy}/D_T \) and relative permeability \( 4 \epsilon_i = [(\epsilon_1 + \epsilon_2)/\epsilon_i]^2 \). In the above formula, we have also introduced the field enhancement factor \( F = E/E_0 \). The dimensionless chirality is thus scaled by a factor that purely depends on the material properties of the twisted van der Waals structure. For screened plasmons, we have

\[
|\mathcal{C}_i/\mathcal{C}_i^0| = \frac{\mu_\mu_0 a q}{\epsilon_i 4 d k_i} \alpha_{xy} F^2 ,
\]

(9)

where \( \alpha_{xy} = \frac{a D_{xy}}{\epsilon_0 c_2} \) is the chiral fine-structure constant. Note that this expression does not depend on the total Drude weight and can be largely enhanced by doping the system. Also the field enhancement is substantially larger such that this platform is likely to yield the strongest chiral near-field without breaking the time-reversal symmetry.

Let us finally comment that Eqs. (8) and (9) would be an artifact unless we relate both \( E \) and \( E_0 \) by a common physical ruler. For that, we imagine that the same plasmonic current intensity \( j_\parallel \) that creates \( |E| \), now shines at a radiative wavevector \( q_r \ll \omega/c \), creating the far-field plane wave of amplitude \( |E_0| \). Then \( F \sim \frac{q_r}{\omega c} \sim \frac{c}{\omega c} \), valid for the typical wavevectors of graphene plasmons.

**Numerical estimates.** The expressions for the local chirality depend on the total and chiral (Hall) Drude weight, shown in Figs. 1 and 2. In Fig. 3, we show the combined chirality \( \chi \) of the system, i.e., \( \chi = D_{xy}/D_T \) that governs the local chirality of unscreened collective charge oscillations and the Poynting vector. Again, the abrupt change at the magic angle is evident (see dashed line).

For large twist angles, we observe in Fig. 3 an odd behavior of the chirality with a maximum around 10. For small twist angles, there is a constant plateau with well-defined chirality for large \( |E_F| \) reflecting the chirality of the lattice. However, around the neutrality this chirality changes sign for \( i = 30 \) and \( D_{xy} \) becomes zero. Notice also that all curves collapse to the same curve.

The analytical expressions of the previous section can be simplified considerably if only the linear term in \( q' a \) is kept and retardation effects are partially neglected by \( q' \rightarrow q \). The Poynting vector of a chiral plasmon then forms the angle \( \tan \theta = -\text{sgn}(z)2q a \) with respect to propagation direction \( q \).\(^{35} \) For \( \chi \approx 10 \) and a chemical potential around \( \mu = 40 \text{meV} \), i.e., a twist angle of \( \theta \approx 2^\circ \) would yield an angle \( \theta \sim 0.5^\circ \) that is formed by \( q \) and \( P \) which should be observable via the plasmon Hall shift.\(^{46} \)

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**FIG. 2.** Left: The total Drude weight in units of \( (e/h)^2 \) for large twist angles with \( i = 10, 15, 20 \) (left) and for small twist angles with \( i = 26, 28, 30 \) (upper right) and \( i = 30, 32, 34 \) (lower right) as function of the chemical potential \( E_F \).
at liquid-nitrogen temperatures.\(^{47}\)

The dimensionless chirality of typical (unscreened) plasmons can be estimated as \(|C/C_0| \approx 10^{-3} \chi F^2\). For screened plasmons, we have \(|C/C_0| \approx D_{xy} F^2\), where \(D_{xy} = D_{xy}(e/\hbar)^2\). We will use these expressions in the following discussion.

**Discussion.** Chirality is an important aspect in life as only one enantiomer of amino acids is present in nature.\(^{48}\) Furthermore, chiral objects can only be distinguished through the interaction with other chiral objects. A prominent example is the circular dichroism of chiral molecules where a different absorption cross section is seen when changing the chirality of the incident light.\(^{49\text{-}53}\) This gives rise to an asymmetry factor denoting the normalised difference between the two absorption cross sections.

Exposing (chiral) organic molecules to chiral, e.g., circularly polarized light (CPL) might lead to modified chemical reactions, but usually the asymmetry factor is small and of the order of \(10^{-3}\) and thus negligible. This is related to the fact that the length scales between the two interacting chiral objects usually differ by several orders of magnitudes, i.e., for CPL \(ak \approx 10^{-3}\) in the optical regime where \(k = 2\pi/\lambda\) denotes the wave number and \(a\) the length scale of the molecule.

There have been proposals to enhance the enantioselectivity in the excitation of chiral molecules by superchiral light.\(^{54}\) Also chiral metamaterials and plasmonics show promising results,\(^{52\text{-}55,56}\) and axial coupling can also be induced in Bernal-stacked bilayer graphene. But with atomically thin two-dimensional crystals, macroscopically large chiral objects can be designed following a bottom-up approach and axial coupling can also be induced.\(^{57}\) The circular dichroism of TBG\(^{27}\) could thus be increased by stacking multiple layers with a definite relative twist angle on top of each other. For \(n\) layers, the intrinsic length scale given by the interlayer separation \(a \sim 3.4\AA\) would be increased by \(a \rightarrow a^* = na\) and the dimensionless ”chirality” \(a^*k\) could reach the order of unity.\(^{27}\) Another way to increase the dimensionless ”chirality” \(ak\) would be to decrease the wavelength of the chiral field which has been pursued here.

Reducing the wavelength of the electromagnetic field is possible using confined plasmonic modes. In fact, the plasmonic wave-length can be considerably reduced in graphene,\(^{58,59}\) which is related to the fact that the Fermi velocity \(v_F\) is two order of magnitude smaller than the speed of light.\(^{6,66}\) Based on this wavelength reductions, there are proposals to enhance two-photon processes\(^{61}\) and to construct ”designer atoms”.\(^{62}\) An alternative, but related approach to alter chemical reactions or to catalyse new ones is to drive the system into the strong light-matter interaction regime.\(^{63,64}\)

Field enhancements in graphene are of the order of \((c/v_F)^2 \approx 10^5\). In the case of twisted bilayer, there is a further reduction of the Fermi velocity\(^1\) and for our conservative estimates, we assume a Fermi velocity renormalisation of \(v^* = v_F/3\) and thus a field enhancement of \((c/v^*)^2 \approx 10^6\). The near field produced by these plasmons would have a chirality more than \(10^3\) times larger than that of circularly polarized light. For screened plasmons,\(^{44}\) the enhancement is even be larger, i.e., \(|C/C_0| \sim 10^4\). This would yield an asymmetry factor in the circular dichroism of order unity and the proposed platform might give rise to unprecedented chemical reactions between chiral molecules that are usually forbidden.

**Summary.** We have introduced a macroscopic quantity that defines the chirality of the electronic subsystem of TBG. This quantity changes sign due to an emergent \(C_4\)-symmetry that occurs when the lowest valence and conduction bands, both endowed with dual \(C_4\)-symmetries, merge and eventually cross at the magic angle. Interestingly, this happens for all wave numbers at approximately the same twist angle. Even more astonishing, we find the absence of chirality on an even larger energy scale which naturally leads to the definition of a magic angle, i.e., the angle for which the chirality of the system vanishes and the two bands closest to the neutrality point merge, measurable in-plane magneto-resistance measurements.

We have further investigated the electromagnetic near-field confined to TBG focusing on its chirality. This is caused without breaking time-reversal symmetry and can, therefore, be useful in the context of catalysing chemical reactions without changing the external conditions, e.g., due to the presence of a magnetic field. We find huge field enhancements paving the way towards “chiral plasmonic chemistry” which is possible for general twisted van der Waals structures\(^{65}\) since the interlayer Moiré coupling induces a chiral response that endows surface plasmons with a chiral character.\(^{31,32}\) Another approach would be to use Berry plasmons,\(^{66,67}\) induced by circularly polarized light that can lead to strong chiralities in the presence of a large Berry curvature induced, e.g., by a small gap in single or bilayer graphene.
Supplemental Material

I. EMERGENT $C_6$-SYMMETRY

The band structure of twisted bilayer graphene can be understood from the hybridization of the Dirac cones of the two carbon layers. Under the twist of the layers, their respective Brillouin zones (BZs) undergo a relative rotation, leading to a mismatch $\Delta K$ of the Dirac nodes at the $K$ points, as shown in Fig. 4. The low-energy bands of the twisted bilayer can be folded then in the two Moiré-BZs represented by the small hexagons in the figure.

In the continuum model, the low-energy bands are obtained separately for each of the two independent Moiré-BZs. Taking for instance the one to the right in Fig. 4, we have the Hamiltonian

$$H = v_F \begin{pmatrix}
0 & -i\partial_x - \partial_y + i\Delta K/2 & V_{AA'}(r) & V_{AB'}(r) \\
-i\partial_x + \partial_y - i\Delta K/2 & 0 & V_{BB'}(r) & 0 \\
V_{AA'}^*(r) & V_{BB'}^*(r) & 0 & -i\partial_x - \partial_y - i\Delta K/2 \\
V_{AB'}^*(r) & V_{AA'}^*(r) & -i\partial_x + \partial_y + i\Delta K/2 & 0
\end{pmatrix}$$

(10)

where $v_F$ is the Fermi velocity of graphene and $V_{AA'}(r), V_{AB'}(r)$ and $V_{BA'}(r)$ are the respective interlayer tunneling amplitudes between regions of stacking $AA, AB$ and $BA$. The Hamiltonian $H_L$ operating in the other Moiré-BZ is similar to $H_R$, with the only difference that the shift $\Delta K$ is replaced by $-\Delta K$ and the momentum $k_x$ is reversed after changing from one graphene valley to the other.

The low-energy bands $\varepsilon_L(k), \varepsilon_R(k)$ obtained from the respective Hamiltonians $H_L, H_R$ do not have in general the same shape, but they are mapped onto each other by the transformation $k_x \to -k_x$. This corresponds to the fact that the lattice of the twisted bilayer remains invariant under a mirror-symmetry transformation about the $y$-axis in real space, accompanied by the exchange of the two carbon layers. In general, each of the bands $\varepsilon_L(k)$ and $\varepsilon_R(k)$ does not have separately such a mirror symmetry under the reversal $k_x \to -k_x$, as shown in Fig. 5. There we also
FIG. 5. Band structure of the first valence and conduction bands for the two valleys at $i = 28$. The change in chirality is clear with respect to the first valence and conduction bands as well as with respect to the two valleys.

Looking at the first valence and conduction bands, however, we see that there is a special twist angle at which the symmetry is enlarged, as the mirror symmetry $k_x \rightarrow -k_x$ becomes realized within each Moiré-BZ of the twisted bilayer (see Fig. 6). The value of such a critical twist angle is in the regime where the first magic angle is usually located, also discussed in the SM of Ref. 36. For the parameters we have used in our calculation (with $t = 2.78$ eV and Fermi velocity $v_F = 4.2$ eV×a, a being the C-C distance, and interlayer coupling $w = 0.11$ eV), the angle at which we find the enlarged symmetry corresponds to the index $i = 30$, in the sequence of commensurate lattices with $\theta_i = \arccos((3i^2 + 3i + 0.5)/\sqrt{3i^2 + 3i + 1})$. The approach to the critical angle is clearly noticed in the contour plots of the low-energy bands, as they are promoted to a $C_6$ symmetry which is in general absent within each Moiré-BZ.

FIG. 6. The band structure of the first two valence and conduction bands for the twist angle at $i = 30$. Notice that the emergent symmetry of $C_6$ is also present in the second valence and conduction band, respectively.

We observe therefore that there is a critical angle at which an enhanced symmetry is realized in the twisted bilayer. This is indeed an emergent symmetry, since it is not implied by the symmetry transformations in the lattice of the bilayer. The location of the critical angle can be taken as a more precise identification of the magic angle, as this is usually referred to a situation where the first valence and conduction bands become flat, while in practice such a flatness is only approximate. In the case of the critical angle here described, the realization of the emergent mirror symmetry is however very precise, as seen in Fig. 7. Moreover, the location of the critical angle has a clear observational counterpart, as remarked in the main text, since it corresponds to the point where the sense of chirality is lost in the twisted bilayer, in accordance with the enhanced symmetry of the low-energy bands. Also approximate symmetry lines at $i = 50$ and $i = 63$ correspond to sign changes in $D_{xy}$.

Let us finally emphasize that chirality cannot be discussed using the band-structure alone, but only in connection with the eigenvalues as is clear from its definition. On the left panel of Fig. 8, we show the density plot of $D_{xy}$ of the first valence band of one valley at $i = 30$ as function of the first Brillouin zone. The black line indicates the sign change of $D_{xy}$, the red and green lines denote the Fermi lines for two characteristic Fermi energies. We observe that for both Fermi energies there is a cancellation of chirality around the Fermi lines.

Interestingly, this cancellation does not only occur within the first valence band, but is also present for higher Fermi energies, i.e., the chirality of the 2nd VB (CB) is almost perfectly cancelled by the 3rd VB (CB) for $i = 30$. For twist
angles close to the magic angle, the interplay between the 2nd and 3rd band further leads to a plateau-like structure. This is demonstrated in the central panels of Fig. 8 where the total $D_{xy}$ as function of the Fermi energy is shown together with the contributions of the several valence and conduction bands. On the right panels of Fig. 8, we finally show that the sum of the 2nd VB (CB) and 3rd VB (CB) always yields a plateau in $D_{xy}$ which are almost equally spaced for commensurate twist angles with $i \sim 30$ and $\Delta D_{xy} = 0.00046 \pm 0.00046$. For the sake of clarity, we plot $D'_{xy}$ for the electron-hole symmetric model that only contains the contribution from 2nd and 3rd valence and conduction bands.

II. NEAR-FIELD PROPERTIES IN 2D SYSTEMS WITH BROKEN TIME-REVERSAL SYMMETRY

Two-dimensional bulk plasmonic properties in systems with time-reversal symmetry cannot easily be distinguished from systems with broken time-reversal symmetry regarding. The reason for that is that the transverse (s-polarised) current sources are suppressed by the fine-structure constant and can thus be neglected in comparison to the longitudinal (p-polarised) current sources. Still, there are quantities which are only non-zero in the retarded regime and these shall be discussed in this work.

Plasmons in two-dimensional systems with broken time-reversal symmetry exhibit a transverse current $j_\perp$ that is associated to the longitudinal current $j_\parallel$. Let us define the complex sources $j_\parallel = j_\parallel e^{iq z} e^{2q' z} e^{-2q z}$ with $q' = \sqrt{q^2 - \mu \epsilon (\omega/c)^2}$ and $e_{q\perp} = e_z \times e_q$. The corresponding real current densities shall be given by $j_\nu = $Re$j_\nu$ with $\nu = \parallel, \perp$.

The associated near-field is given by $A_\nu = -D_\nu j_\nu$ where $D_\nu$ is the longitudinal ($\nu = \parallel$) or transverse ($\nu = \perp$) photonic propagator, respectively.\(^{45}\) The parallel current $j_\parallel$ will thus give rise to a longitudinal field and the perpendicular current $j_\perp$ to a transverse field. With the total gauge field $A = A_\parallel + A_\perp$ and $E = i\omega A$ and $B = \nabla \times A$, we thus
obtain the following expression:

\[
E = i\omega \begin{pmatrix} -d_l j_l & -d_l j_\perp & -i\text{sgn}(z) \frac{q'}{q} d_l j_l \\ -d_l j_l & -d_l j_\perp & -i\text{sgn}(z) d_l j_l \\ -i\text{sgn}(z) \frac{q'}{q} d_l j_\perp & -i\text{sgn}(z) d_l j_\perp & -iqd_l j_\perp \end{pmatrix} e^{iqx} e^{-q'|z|}
\]

(11)

\[
B = \begin{pmatrix} -\text{sgn}(z) q'd_l j_\perp & -\text{sgn}(z) \frac{k_0^2}{q} d_l j_l & -iqd_l j_\perp \\ -\text{sgn}(z) q'd_l j_\perp & -\text{sgn}(z) \frac{k_0^2}{q} d_l j_l & -iqd_l j_\perp \end{pmatrix} e^{iqx} e^{-q'|z|},
\]

(12)

where \(d_l = \frac{q'}{2\epsilon_0 \omega^2}, d_t = -\frac{\mu\mu_0}{2q'}, k_0 = \omega/c\) and \(e_q = e_x\).

A. Optical momentum, spin, angular momentum, and helicity

The local energy density \(w\), "complex" Poynting vector \(\Pi\) (the Poynting vector is defined as \(P_{Poy} = \Re \Pi\)), momentum \(P\), spin \(S\), and helicity \(H\) of a monochromatic electromagnetic wave can be defined as follows:\(^{37–40,43}\)

\[
w = \frac{\epsilon\epsilon_0}{4} E^* \cdot E + \frac{1}{4\mu\mu_0} B^* \cdot B
\]

(13)

\[
\Pi = \frac{1}{2\mu\mu_0} E^* \times B
\]

(14)

\[
\omega P = \frac{\epsilon\epsilon_0}{4} \Im E^* \cdot (\nabla)E + \frac{1}{4\mu\mu_0} \Im B^* \cdot (\nabla)B
\]

(15)

\[
\omega S = \frac{\epsilon\epsilon_0}{4} \Im E^* \times E + \frac{1}{4\mu\mu_0} \Im B^* \times B
\]

(16)

\[
\omega H = -\frac{1}{2\mu\mu_0} \Im E^* \cdot B
\]

(17)
For the above current density, this yields the following local properties with $\mathbf{q} = q\mathbf{e}_z$:

$$
\begin{align*}
\mathbb{w} &= \frac{\mu_0 j^2}{8 k_0^2} q^2 \left( 1 + \frac{j^2}{j^2} \frac{k_0^2}{(q')^2} \right) e^{-2q'|z|} \\
\Pi &= \frac{\mu_0 j^2 q \omega}{8 k_0^2} \left( 1 + \frac{j^2}{j^2} \frac{k_0^2}{(q')^2} sgn(z) 2 \frac{j^2}{j^2} \frac{k_0^2}{(q')^2} \right) e^{-2q'|z|} \\
\mathbf{P} &= \frac{q}{\omega} w e_q e^{-2q'|z|} \\
\mathbf{S} &= -sgn(z) \frac{\mu_0 j^2 q}{8} \frac{1}{\omega} \frac{q^2}{q'} \left( \frac{2j^2}{j^2} \frac{k_0^2}{(q')^2} \left( 1 + \frac{j^2}{j^2} \frac{k_0^2}{(q')^2} \right) \right) e^{-2q'|z|} \\
\mathcal{H} &= -sgn(z) \frac{\mu_0 j^2 q}{4} \frac{1}{q'k_0^2} e^{-2q'|z|}
\end{align*}
$$

B. Ellipticity

We can also define the ellipticity of the electric and magnetic field as follows

$$
\begin{align*}
\mathcal{E}_E &= \Im \frac{E^* \times E}{E^* \cdot E} \cdot \mathbf{P}_{Poy}, \\
\mathcal{E}_B &= \Im \frac{B^* \times B}{B^* \cdot B} \cdot \mathbf{P}_{Poy}, \\
\mathcal{E} &= \Im \frac{\epsilon \epsilon_0 E^* \times E + \frac{1}{\mu_0} B^* \times B}{\epsilon \epsilon_0 E^* \cdot E + \frac{1}{\mu_0} B^* \cdot B} \cdot \mathbf{P}_{Poy},
\end{align*}
$$

with $\mathbf{P}_{Poy} = \Re \Pi$. Interestingly, we find $\mathcal{E}_E = \mathcal{E}_B$ with

$$
\mathcal{E}_E = -sgn(z) \frac{\mu_0 j^2 q^2}{4} \frac{\omega}{q'} e^{-2q'|z|}.
$$

Even though the electric and magnetic fields are coupled, we can assign the same ellipticity to each sector. Consequently, we also have $\mathcal{E} = \mathcal{E}_E$. Furthermore, there is a relation between the ellipticity and helicity with $\mathcal{E} = \omega \mathcal{H}$.

C. Chirality

The local chirality for a real electromagnetic field in a dielectric medium ($\epsilon$, $\mu$) is defined by

$$
\mathcal{C} = \frac{\epsilon \epsilon_0}{2} \mathbf{E} \cdot (\nabla \times \mathbf{E}) + \frac{1}{2\mu_0} \mathbf{B} \cdot (\nabla \times \mathbf{B}).
$$

Note that a finite contribution to the chirality only comes from the scalar product involving both, the longitudinal and the transverse field component which justifies the denomination of this conserved quantity. The resulting fields, therefore, display the following chirality:

$$
\mathcal{C} = -sgn(z) \frac{\mu_0 j^2 q^2}{4} \frac{\omega}{q'} e^{-2q'|z|}.
$$

The optical chirality is associated to a flux of chirality related by the usual continuity equation $\partial_t \mathcal{C} + \nabla \cdot \mathbf{F} = 0$ (in the absence of material currents) which is locally defined as

$$
\mathbf{F} = \frac{1}{2\mu_0} \left( \mathbf{E} \times (\nabla \times \mathbf{B}) - \mathbf{B} \times (\nabla \times \mathbf{E}) \right).
$$
For the chiral plasmon, this gives

$$\mathcal{F} = -\text{sgn}(z)\frac{\mu_0 \omega q}{8} e^{-2q' |z|} \left[ 2j_{\parallel} j_{\perp} e_q + j_{\parallel}^2 \left( \frac{q'}{k_0} \right)^2 \left( 1 + \frac{j_{\perp}^2}{j_{\parallel}^2} \frac{k_0^2}{q'} \right) e_{q_{\perp}} \right].$$

(30)

Notice that we have $\mathcal{F} = \omega^2 \mathbf{S}$ which gives rise to a conserved “spin-density” $\mathbf{S} = C/\omega^2$. We also have $\mathcal{H} = C/k_0^2$.

Both relations are general and obtained by noting that

$$\mathcal{C} = \frac{\epsilon_0}{2} (\mathbf{B} \cdot \partial_t \mathbf{E} - \mathbf{E} \cdot \partial_t \mathbf{B}) = k_0^2 \mathcal{H},$$

(31)

$$\mathcal{F} = \frac{\epsilon_0}{2} \mathbf{E} \times \partial_t \mathbf{E} + \frac{1}{2\mu_0} \mathbf{B} \times \partial_t \mathbf{B} = \omega^2 \mathbf{S}.$$  

(32)

The chirality is also linked to the ellipticity with $\mathbf{E} = \omega \mathbf{C}/k_0^2$.

### III. Near-Field Properties in 2D Systems with Time-Reversal Symmetry

In the previous Sec. II, we have analyzed the near-field response in one layer for which time-reversal symmetry is explicitly broken. This treatment can be extended to a bilayer with the two layers located at $z_1 = a/2$ and $z_2 = -a/2$. An alternative approach considering an effective single electro-magnetic sheet can also be found in Sec. IV.

Let us now explicitly consider two different dielectrics $\epsilon_i, \mu_i$ with $i = 1, 2$ in the two half-planes $|z| > a/2$. The electromagnetic field is usually characterized by the local density $w_i$ and the local Poynting vector $\mathbf{P}_i$ of each half-plane. In the limit $aq'_i \ll 1$ with $q'_i = \sqrt{q^2 - \mu_i \epsilon_i (\omega/c)^2}$, we obtain the following expressions:

$$w_i = \frac{\mu_i \mu_0 j_0^2}{2} \frac{q^2}{k_i^2} \left( 1 + \tilde{j}_{\perp}^2 \right) e^{-2q'_i |z|},$$

(33)

$$\mathbf{P}_i = \frac{\mu_i \mu_0 j_0^2}{2} \frac{q \omega}{k_i^2} \left[ \left( 1 + \tilde{j}_{\perp}^2 \right) e_q + \text{sgn}(z) \frac{j_{\parallel}^2}{j_{\perp}^2} q'_i a e_{q_{\perp}} \right] e^{-2q'_i |z|},$$

(34)

where we defined $\tilde{j}_{\perp} = [1 + j_{\parallel}^2 (k_i ^2/q^2)^2]^{1/2}$ and $k_i = \omega/c_i$ with $c_i = c/\sqrt{\mu_i \epsilon_i}$ the speed of light of the dielectric medium.

For the near-field chirality of TBG, we have (as stated in the main text)

$$C_i = -\frac{\mu_i \mu_0}{2} a q'_i j_{\parallel} j_{\perp} e^{-2q'_i |z|},$$

(35)

$$\mathcal{F}_i = -\frac{\mu_i \mu_0 \omega q}{2} q'_i \left[ 2j_{\parallel} j_{\perp} q'_i a e_q + \text{sgn}(z) \frac{j_{\parallel}^2}{j_{\perp}^2} \frac{(q'_i)^2}{k_i^2} \left( 1 + \tilde{j}_{\perp}^2 \right) e_{q_{\perp}} \right] e^{-2q'_i |z|}.$$  

(36)

Notice that the Poynting vector as well as the chirality flux contain a non-trivial transverse component which could be chosen arbitrarily without violating the continuity equation. The local definition of $\mathbf{P}$ and $\mathcal{F}$ thus goes beyond the transport properties.$^{33}$

### IV. Chirality in Magneto-Electric Sheets

For chiral plasmons, the classical picture of field sources in vacuum are in-plane, longitudinal current and magnetic moment densities, written as

$$2j(r, t) = j_0 e^{i q \cdot r} e^{-i \omega t} + \text{c.c.}$$

$$2m(r, t) = a n \cdot j_0 e^{i q \cdot r} e^{-i \omega t} + \text{c.c.},$$

(37)

with $j_0 \parallel q$, and where $n \times$ is a material constant that quantifies the parallel magnetic moment following the current. $a$ represents an intrinsic length (interlayer distance for twisted bilayer), introduced to make $n \times$ dimensionless.

The fields associated with these sources, $\mathbf{E}_{j,m}$ and $\mathbf{B}_{j,m}$, can be calculated explicitly but, to show that $\dot{C}$ is non zero, and the generality of the argument, it suffices to realize that: i) only the crossed terms ($j, m$) contribute to $\dot{C}$ on symmetry (parity) grounds, ii) the term from $\mathbf{E}_m \mathbf{B}_j$ is smaller than the term $\mathbf{E}_j \mathbf{B}_m$ by factors of $(\frac{a}{q})^2$, iii) the
The electric and magnetic fields of an electric dipole can be read from the magnetic and electric counterparts of a magnetic dipole.

The last point and the relation between current and dipole density, \( j = \partial_t \mathbf{p} \), allow us to write

\[
B_m = i\omega\epsilon_0\mu_0\mathbf{p}A_j,
\]

and finally

\[
C = k_0^2\frac{\epsilon_0}{4}(\mathbf{n} + \mathbf{n}^*)a|\mathbf{E}_j|^2.
\]

We recall that \( \mathbf{E}_j \) is the electric field associated with the longitudinal plasmon current and, for the near field, one could safely take the instantaneous approximation for it. Its explicit expression in terms of \( \mathbf{j}_0 \) will lead to the standard exponential decay \( e^{-2q|z|} \) of near fields.

As a final remark, one notices that a real value of \( \mathbf{n} \) is required for finite \( \mathbf{C} \). This implies that the magnetic moment has to have a component in phase with the plasmon longitudinal current and, therefore, the magnetic dipole density is in quadrature with the electric dipole density. This is precisely the condition for an atomic transition to be chirally active, so the whole picture is consistent. Furthermore, although the formalism is tailored to layered systems, the generality of the arguments implies that a chiral near field should exist whenever the sources comply with the previous requirement of parallel and in quadrature electric and magnetic moments.
For the tight-binding model, it generalizes to
\[ D_{xy}(\mu) = \frac{1}{\pi} \sum_{k,n} e^x \cdot (\mathbf{j}_{k,n} \times \mathbf{j}_{k,n}) \delta(\epsilon_{k,n} - \mu), \]
where \( \mathbf{j}_{k,n} \) resembles the total current including the interlayer current \( \mathbf{j}_{k,n} \), see Ref. 31.

See Supplementary Material for more details with additional analytical and numerical results, which includes the Refs. [36-40].

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