

Non-local chirality in twisted multilayer

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We briefly review basic concepts and requirements for chiral response in multilayer moiré systems.

I. CURRENT STATUS

Chirality is used in many different contexts to label physical phenomena that break reciprocity. However, etymologically, it comes from the Greek word for “hand” and is thus related to a three-dimensional object whose mirror image does not match the original. In reverse, this means that purely two-dimensional systems cannot be genuinely chiral.

Chiral objects define two enantiomers which are identical in all scalar properties such as density or eigenfrequencies. Nevertheless, in their interaction with other chiral objects, opposite enantiomers can be detected. Circularly polarized (CP) light can thus distinguish left or right-handed samples by measuring the different absorption cross-sections of left and right CP light giving rise to circular dichroism (CD) [1]. Since linearly polarized light is the superposition of left and right CP light, there will also be a rotation of the polarization plane. This has been observed in neutral twisted bilayer graphene (TBG) for certain resonant frequencies related to transitions around the van Hove singularities [2].

The observation of CD in TBG is remarkable since Maxwell’s equations are reciprocal in purely two-dimensional systems. Optical activity can thus only be observed by the breaking of certain symmetries, and typically time-reversal or rotational symmetry are broken [3]. In TBG, both symmetries are conserved and the CD is due to the breaking of the two-dimensionality of the system as the two layers are separated by distance $d_{\perp} = 3.4\text{\AA}$. In fact, the chiral response is due to the *non-local* correlation between the current-density of layer 1 in say x -direction and the current density of layer 2 in the (transverse) y -direction:

$$\chi^{chiral}(\omega) = -\frac{i}{\hbar} \int_0^{\infty} dt e^{i\omega t} \langle [j_x^1(t), j_y^2(0)] \rangle, \quad (1)$$

where $j_{\nu}^{\ell}(t)$ is the ν -directed current operator ($\nu = x, y$) in layer ℓ in the interaction picture, and $\langle \cdot \rangle$ is the

equilibrium average. Eq. 1 provides the average current in layer 1 due to the electric field in layer 2, and could be interpreted as the system response to the “gradient” of the electric field along the third dimension [4].

The chiral response only depends indirectly on the distance between the two layers, d_{\perp} , via the equilibrium average. However, typical chiral observables are directly proportional to d_{\perp} and thus depend on $d_{\perp} \chi^{chiral}$ [5].

The non-local response between perpendicular current-directions of different layers is not the only requirement to observe CD, because for electron-hole symmetric systems the chiral response is identically zero at charge neutrality. Being only due to small electron-hole symmetry breaking, electron-like and hole-like transitions are not compensated and the enhanced density of states around van Hove singularities can give rise to an observable CD [6, 7]. In fact, two main resonances are observed in Ref. [2], related to transitions around two different van Hove singularities.

II. CHALLENGES AND OPPORTUNITIES

To calculate the CD, one needs to go beyond the dipole approximation $e^{ikz} \approx 1 + ikz$, again emphasizing that chirality is a three-dimensional phenomena. This introduces the dimensionless scale ka , where a is the typical extension of the chiral object and $k = 2\pi/\lambda$ the wave number of light. This scale is usually small making chiral effects hard to use in typical nanoscale devices.

Field-theoretically, the chirality of an electromagnetic field with electric field \mathbf{E} and magnetic field \mathbf{B} in a dielectric medium with relative dielectric permittivity ϵ and magnetic permeability μ is defined by

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

which can further be related to a chiral flux via a continuity equation [1]. For CP light, the chirality is proportional to its frequency ω and intensity $|\mathbf{E}_0|^2$. For fixed wavelengths, the chirality can thus only be modified by

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changing the amplitude which can be achieved by confining light. In fact, decay into evanescent modes gives rise to fluorescence quenching which has been used to detect chiral molecules with the help of TBG [8].

Also, surface-plasmon polaritons could enhance the chiral coupling and may enhance the chirality up to 4000 times the one of corresponding propagating CP light [9]. Cavities composed of TBG should thus offer tremendous opportunities as plasmon-induced near-field chiralities may enhance photocatalytic processes for enantiomer-selectivity.

Quasi two-dimensional moiré structures can host plasmons which are inherently chiral [5]. Especially, many organic molecules are intrinsically chiral mainly due to the chemical structure of carbon with its 4 valence electrons. In fact, all amino acids are chiral as only glycine is achiral. Moreover, the RNA aptamer or Spiegelmer is chiral as well as hexahelicene and pentahelicene. All these may be synthesized in an enantiomer-selective way giving rise to new functionalities.

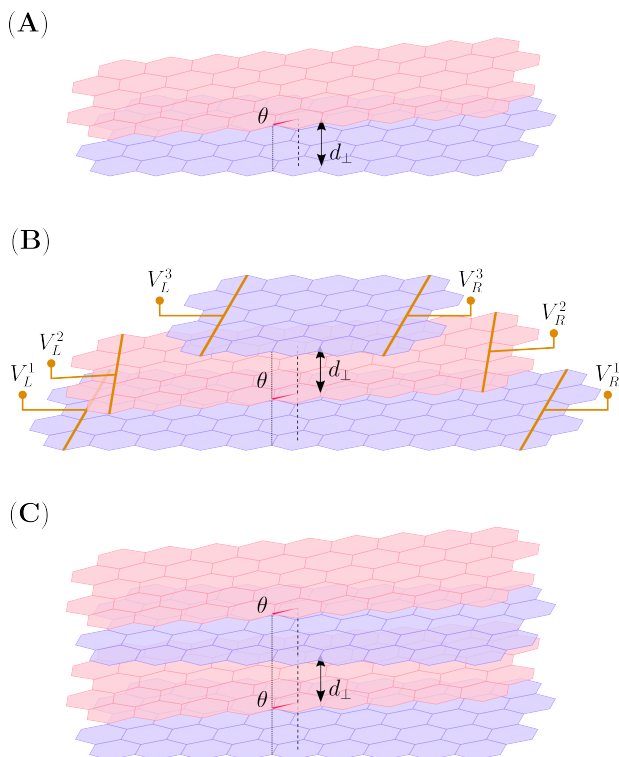


FIG. 1. Multilayer moiré systems with $N = 2$, $N = 3$, and $N = 4$ layers. The structures with alternating twist angles and an even layer number are inherently chiral, whereas those with an odd layer number are achiral as they display a mirror plane at the central layer.

III. FUTURE DEVELOPMENTS TO ADDRESS CHALLENGES

The crucial requirement for the chiral response in moiré systems is the spatial separation between the layers and thus the non-local correlations between the sheet currents. However, increasing the layer distance would result in weakening the quantum mechanical coupling. Hence, there is a trade-off between optimal coupling and large layer separation, probably best met by transition-metal dichalcogenides such as WSe_2 .

Another route to enhance the inherent chirality is to stack several van-der-Waals materials on top of each other [10, 11]. The chiral response of multilayers with width d should thus be increased by the geometric factor $d/d_{\perp} = n$ where n is the number of layers. However, the moiré unit cell grows exponentially with the number of layers even for commensurable twist angles, and for incommensurable structures it is difficult to obtain the electromagnetic response [12].

For the special case of alternating twisted multilayers, the analysis can be considerably simplified as it can be mapped to independent twisted bilayers and one single layer in case of an odd number of layers [13]. In Fig. 1, the multilayer moiré systems with $N = 2$, $N = 3$, and $N = 4$ layers are shown. The structures with an even layer number are inherently chiral, whereas those with an odd layer number are achiral as they display a mirror plane at the central layer. Nevertheless, even for $N = 3$ the non-local “chiral” correlations that involve the perpendicular current densities of adjacent layers give rise to vertical gradients of the magnetic moment as a response to an electric in-plane field [14]. This layer non-locality in nominally “achiral” systems could be detected by layer-discriminated contacts, as shown in Fig. 1 (B) and also discussed in Ref. [15].

Let us note that we can estimate the chiral response for moiré systems with *arbitrary* twist angles between the layers using the decoupling procedure proposed in [13]. For that, we rely on the fact that decreasing (increasing) twist angles can be mimicked by properly increasing (decreasing) the interlayer tunnel amplitude [16]. Neglecting effects of non-commensurability, we can thus map any multilayer moiré system onto a set of independent twisted bilayers and one single layer in case of an odd number of layers.

IV. CONCLUDING REMARKS

The non-local coupling between sheet-currents flowing the perpendicular directions gives rise to a chiral response for neutral multilayer moiré systems without a mirror plane and slight electron-hole asymmetry. Also for multilayer moiré systems with a mirror plane, non-local “chiral” correlations can be detected by layer-discriminating contacts. These effects can

be considerably enhanced by non-radiative (plasmonic) modes leading the path to chiral cavities composed of multilayer moiré systems with improved enantiomer-selective capabilities. The functionality can be further optimized by tuning the twist angles and doping levels.

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