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Symmetry frustration and chiral electronic order in transition-metal dichalcogenides

1. **Origin of chirality in transition-metal dichalcogenides**
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   arXiv:2312.11979
2. **Observation of giant circular dichroism induced by electronic chirality**
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   arXiv:2312.11961, Phys. Rev. Lett. 133, 126402 (2024)

Recommended with a Commentary by Yuan Li, International Center for Quantum Materials, Peking University

Chirality is a key feature of geometric objects that lack improper rotational symmetry (*i.e.*, a combination of a rotation and a reflection, represented by orthogonal matrices with determinant -1). It is tempting to think about the experience of an electron in chiral materials by taking an analogue to magnetic field effects such as the Lorentz force, which bends an electron's motion by the right-hand rule. In loose terms, when combined with a sense of motion such as that of an electric current or light propagation, a chiral material can produce effects mimicking magnetization [1].

Chiral crystal structures are not rare. Among the 32 (230) crystallographic point (space) groups, 11 (65, known as Sohncke groups) describe the symmetry of chiral crystals. In most familiar examples such as tellurium, selenium, α-quartz, sodium chlorate *etc.*, the chiral structure is formed at a high temperature, directly from a molten state or by chemical reaction. A notable yet rare instance of low-energy scale chirality formation occurs in the charge-density waves (CDWs) of $1T$-TiSe$_2$ [2]. This discovery has spurred targeted experiments aiming to distinguish and manipulate the chirality on a macroscopic scale [3], as well as investigations of chiral electronic order in related materials [4–7].

An unsettled question remains: What makes the CDW order chiral in $1T$-TiSe$_2$? The order is known to have a triple-**q** character, which means that the charge modulations are a sum of three sinusoidal waves with the wave vectors rotated by 120°. A phenomenological way to obtain chirality from this is by letting the sinusoidal waves have relative phase shifts

[2, 8]. In addition, the transverse displacement of Ti atoms with respect to the wave vectors invites an interpretation of the CDWs as orbital order [8]. However, the phenomenological theory predicts that the relative phase shifts only occurs when the CDW order parameters become sufficiently large. This implies that there ought to be a separate chirality transition at a somewhat lower temperature [8] than the main, achiral CDW transition, yet observation of such a separation in diffraction experiments is a subject of debate [9–11].

In the first paper, Kim *et al.* demonstrate through group theory that scalar charge modulations and vector lattice displacements correspond to distinct irreducible representations (IRs) of $1T$-TiSe$_2$'s symmetry. When the displacements are further constrained to be transverse to the modulation wave vectors, as supported by the corresponding phonons' X-ray scattering cross sections, they become a complete stranger to charge-density modulations on the ions (Fig. 1). As such, the simultaneous development of both types of modulations requires further lowering the symmetry, which results in a chiral structure. Relative phase shifts between the sinusoidal waves are no longer explicitly required. The broken symmetries are confirmed in the paper by the appearance of extra elastic X-ray scattering peaks and by the splitting of certain phonon modes seen with Raman spectroscopy.

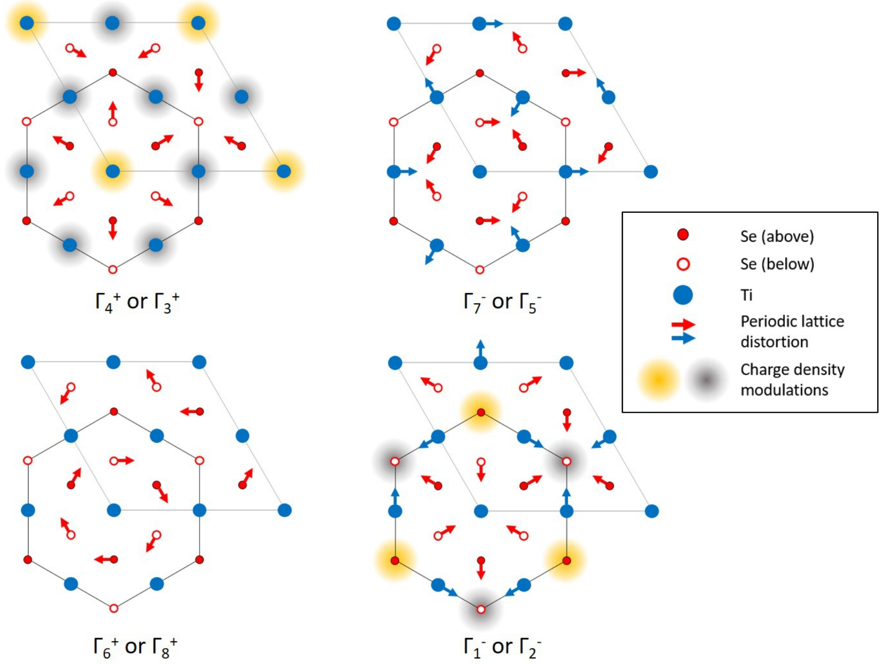

Figure 1: Isotropic basis functions including planar lattice distortion and charge-density modulations in a 2 × 2 unit cell of a single layer. They are back-folded from the L (Γ_4^+, Γ_6^+, Γ_7^-, and Γ_1^-) and M (Γ_3^+, Γ_8^+, Γ_5^-, and Γ_2^-) points of the original Brillouin zone, which differ only in the inter-layer arrangement. Each of the basis functions can be decomposed into three equal single-**q** components shown in the first paper. In principle, the sign of the charge-density variation (yellow and grey shading) and the directions of Ti and Se displacement can be separately reversed without violating the symmetry. Because transverse displacement of Ti (as in Γ_7^-) shares no common symmetry with the charge, they are unable to "complete" a free-energy term at the linear order. As a result, additional symmetry breaking is expected.

In the second paper, Xiao *et al.* use resonant X-ray diffraction to show that the intensity of forbidden Bragg peaks that emerge with the charge order is sensitive to the choice of left- vs. right-hand circular polarization of the incoming photons. The circularly dichroic intensities further depend on the azimuthal scattering geometry, in a fashion that matches calculation results based on a specific form of distorted structure which supports chiral orbital order of the localized Ti $3d$ electrons. Unlike modulations in the total charge density, modulations in the orbital occupation share the same symmetry as atomic displacement, so they naturally occur together. In both papers, only a single thermal phase transition is observed, consistent with the expectation that the charge, orbital, and lattice degrees of freedom all participate in the symmetry breaking in a frustrated manner.

In correlated materials, some of the outstanding mysteries are associated with various symmetry breaking revealed by different experimental probes (an example of this is the pseudogap phase of high-T_c cuprates). Could some of the symmetry breaking be understood as a consequence of frustration between other symmetry-breaking tendencies, just as the chirality formation in $1T$-TiSe$_2$? In addition to crystallographic symmetries, more experimental work is needed to explore possible connections between chiral order and time-reversal symmetry breaking, such as for the chiral CDWs in kagome materials [7, 12]. Inversely, it has been conceived that the formation tendency of triple-$\mathbf{q}$ magnetic order with spin chirality [13, 14] may give rise to a novel form of vestigial charge order [15] that preserves the time-reversal symmetry. Circularly polarized photon scattering experiments may generally be expected to elucidate such electronic phases and their interplay. As we continue to explore the complex interactions of lattice, charge, orbital, and spin degrees of freedom in materials like $1T$-TiSe$_2$, further experimental and theoretical work will be essential. Understanding these interactions and the symmetry interplay could unveil new aspects of phase behavior and emerging phenomena.

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