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Crystalline chirality and interlocked double hourglass Weyl fermion in polyhedra-intercalated transition metal dichalcogenides

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Abstract

Introducing crystalline chirality into transition metal dichalcogenides (TMDs) has attracted much attention due to its modulation effect on optical properties and the potential to reveal new forms of electronic states. Here, we predict a number of chiral materials by intercalating polyhedra into TMD lattices, finding a type of double hourglass Weyl fermion interlocked with crystalline chirality. The best candidate RhV_3S_6 ($P6_322$) possesses the largest hourglass energy window of ~ 380 meV, as well as strong optical circular dichroism (CD) in the infrared regime, both of which are tunable by external strains. The chirality is originally induced by the configuration of intercalated polyhedra and then reduced by the rotational atomic displacements triggered by intercalation, as indicated by CD calculations. Our study opens the way of designing chiral materials with spin-split double hourglass Weyl fermions *via* structural unit intercalation in achiral crystals for future chiral-functionalized optoelectronic and spintronic devices.

Introduction

Transition metal dichalcogenides (TMDs) are the subject of intense interest due to their remarkable electronic and optical properties, such as their direct bandgap in a monolayer limit¹, strongly bound excitons and trions^{2,3}, and chiral optical selection rules^{4,5} due to spin–valley locking degrees of freedom. Benefitting from these unique properties, especially the chiral selection rules, TMDs have acted as an ideal class of materials for optoelectronic and spintronic manipulation, becoming increasingly studied for valleytronic devices⁶. As the chiral optical selectivity^{4,5} of the K and K' valleys have

opposite signs in TMDs without crystalline chirality, external chirality has been applied to tune the valleytronic property of TMDs^{7–10}. Valley polarized photoluminescence of TMDs such as MoS_2 can be tailored through nearfield interactions with plasmonic chiral metasurfaces⁸. The coupling of valley excitons in monolayer WS_2 with chiral surface plasmons has also been demonstrated¹⁰, which occurs at room temperature and persists for a long lifetime. Most recently, external crystalline chirality was induced in MoS_2 nanostructures *via* the surface modification of chiral ligands^{11,12}. The coupling of external chirality with intrinsic chiral optical selectivity significantly enriches the phenomena in the TMD material family.

Weyl fermions in TMDs^{13–17} have also attracted much attention in light of their extremely large magnetoresistance for potential applications in magnetic sensors and memory^{18,19}. The Weyl points in these achiral TMD Weyl semimetals^{13–15} are paired with opposite chirality. Therefore, coupling external net chirality to the chirality of Weyl fermions could further advance the study of

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TMD-based materials and potentially open the way of finding new fermions, such as the Kramers-Weyl fermions²⁰ that are commonly found in nonmagnetic chiral crystals when Kramers theorem-forced band degeneracy appears near the Fermi level (E_F). The recently found new form of fermion, namely, the hourglass fermion^{21–24}, is also closely related to the Weyl fermion, which, however, has not yet been found in TMDs. This class of new fermions was originally predicted on the surface of nonsymmorphic crystal KHgSb²² and then widely found in the electronic structures of nonsymmorphic materials, including three-dimensional ReO_2 ²⁵, Al_3FeSi_2 ²⁶, Ag_2BiO_3 ^{27,28}, AgF_2 ²⁹, and Tl_3PbBr_5 ³⁰, as well as the hypotheticalal two-dimensional structure Bi/Cl–SiC(111)³¹. The predicted hourglass fermion on the surface of KHgSb²² has been observed in experiments *via* angle-resolved photoemission spectroscopy³². However, this type of hourglass-like electronic structure usually appears as single sets. Pairs of hourglass Weyl fermionic electronic structures have not been discussed before, which is plausible, as their splitting and annihilation could be associated with a physical quantity, such as crystalline chirality.

In this paper, we search for TMD-based chiral materials with double hourglass Weyl fermions by polyhedral intercalation in a hexagonal lattice. Based on the stability and electronic property evaluation in the framework of density functional theory (DFT)^{33,34}, we found a few intercalated chiral TMDs with pairs of hourglass fermionic electronic structures around E_F and identified RhV_3S_6 ($P6_322$) as the best candidate with the largest hourglass window of ~ 380 meV. The splitting and spin polarization of the double hourglass fermion was interlocked with the crystalline chirality. We evaluated the optical circular dichroism (CD) for RhV_3S_6 and found that when the rotational atomic displacements around the chiral axis triggered by polyhedral intercalation were increased (decreased), the CD response was reduced (enhanced), suggesting that crystalline chirality was originally induced by polyhedral intercalation and then reduced by rotational atomic displacements. Furthermore, the energy window of the double hourglass fermion and optical CD could be effectively tuned by external strains. Our study opens the way for designing TMD-based materials with interlocked crystalline chirality and double hourglass Weyl fermions, with potential applications in future optoelectronic and spintronic devices.

Methods

Electronic structure calculation

All electronic structures were calculated by DFT³⁵ as implemented in the Vienna ab initio simulation package^{33,36}. The projector-augmented wave pseudopotentials³³ with the exchange-correlation functional of

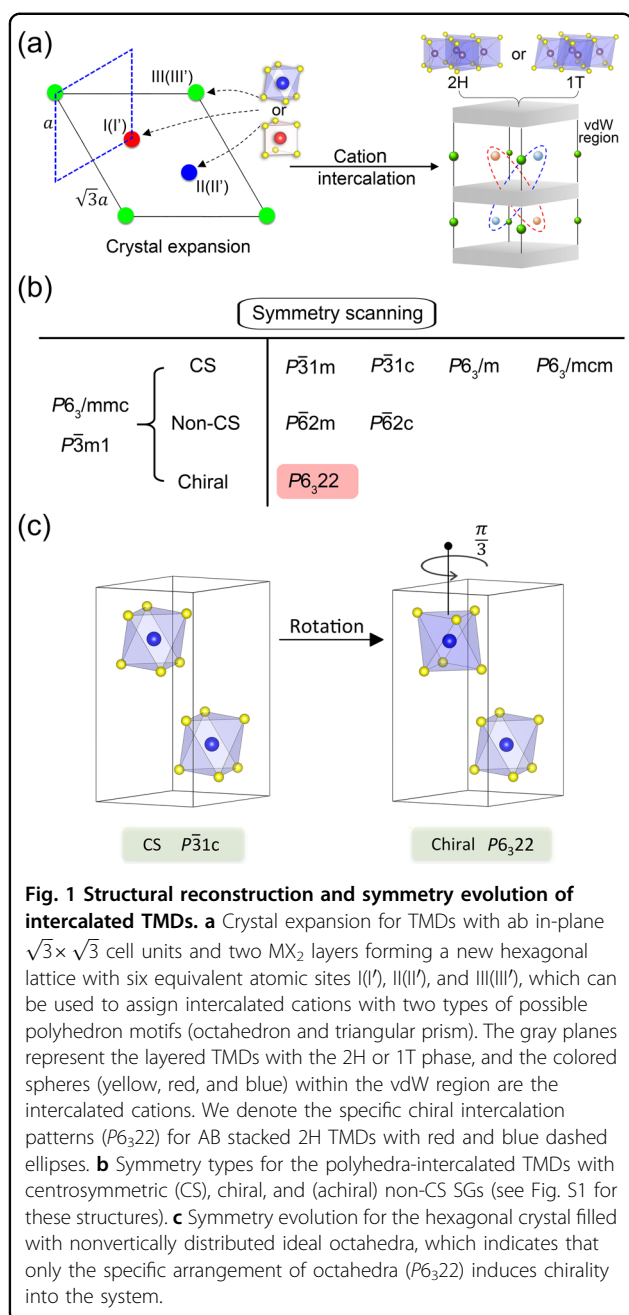
Perdew–Burke–Ernzerhof³⁴ were adopted. The cutoff energy for the plane waves was selected as 520 eV, and the Brillouin zone was sampled on $10 \times 10 \times 4$ Monkhorst-Pack³⁷ k -point meshes. The lattice constants and atomic positions were fully relaxed when the absolute total energy difference between two successive loops and the Hellmann–Feynman force on each atom were $< 10^{-6}$ eV and 5×10^{-3} eV Å^{−1}, respectively, using the conjugate gradient algorithm. The band structure from the hybrid functional (HSE06)^{38,39} was obtained for comparison with the conventional DFT result. Perturbation was added to the pseudopotential to address the spin–orbit coupling (SOC) effect. The spin state of the electron wavefunction was evaluated by projecting the calculated wavefunction on the spin and orbital basis of each atomic site, which generated spin-projected band structures with dense k -meshes in the Brillouin zone. Strain was applied for the deformation calculation by inducing lattice shrinkage and expansion for tensile and compressive situations.

Circularly polarized evaluation

The circularly polarized optical absorption was the difference between the absorption of left- and right-handed lights for all k points within the valence bands and the conduction bands, and these could be normalized and evaluated from the conventional DFT-calculated ω frequency-dependent imaginary part of the dielectric function with the random phase approximation (RPA) and Fermi golden rule^{40–42}:

$$\begin{aligned} \epsilon_{\alpha\beta}^{(2)}(\omega) = & \frac{e^2 h^4}{4\pi^2 \Omega \omega^2 m_e^2} \lim_{q \rightarrow 0} \sum_{c,v,k} 2w_k \delta(E_{ck+q} - E_{vk} - \omega) \\ & \times \langle u_{ck} | i\nabla_\alpha - k_\alpha | u_{vk} \rangle \langle u_{ck} | i\nabla_\beta - k_\beta | u_{vk} \rangle^* \end{aligned} \quad (1)$$

where Ω is the volume of the primitive cell, e is the elementary charge, h is the Planck constant, ω represents the angular frequency, m_e is the electron rest mass, c and v indicate the conduction and valence band states, respectively, E is the eigenenergy of the wavefunction, \mathbf{q} stands for the Bloch vector of the incident wave, u is the wavefunction, w_k denotes the k -point weights that sum to 1 and the factor 2 before it accounts for the fact that degenerating spin has been considered, and ∇ is the momentum operator, in which the sum over momentum \mathbf{k} is conducted over the full 3D space. The optical CD is obtained by decomposing the circularly polarized part from the total absorption spectrum, as implemented in the PWmat code^{43,44}. The circularly polarized response for RhV_3S_6 was determined with the RPA-GGA approach. A planewave basis set was employed at a cutoff energy of 80 Ry, and a total of 212 bands were included to ensure convergence of all computed quantities. A very dense k -point mesh of $14 \times 14 \times 6$ over the reducible hexagonal Brillouin zone was sampled in our calculations.



Thermodynamic and kinetic stability assessment

Usually, we can construct a set of candidate structures (containing their compositions) that exist in crystal databases, e.g., ICSD⁴⁵ or Material Project⁴⁶, compute their total energies, and then screen this list for the lowest-energy structure. To determine the thermodynamic stability of AB_3C_6 compounds, we considered all of its possible disproportionation channels to the competing phases (elemental phases, binaries and ternaries) by solving a set of inequalities in multidimensional spaces of chemical potentials ($\Delta\mu_A$, $\Delta\mu_B$, and $\Delta\mu_C$), which were

presented in detail in our previous work⁴⁷. The most stable cation intercalation position was confirmed through dividing the vdW planes of the layered TMDs with a dense mesh, globally detecting the symmetry evolution and total energy landscape of the system. To further evaluate the dynamic stability, we calculated the phonon dispersion curves using the finite-displacement approach as implemented in the Phonopy code⁴⁸. The phonon frequency was derived from crystal forces originating from displacements of certain atoms in a $2 \times 2 \times 2$ supercell for RhV_3S_6 with $P6_3/22$ symmetry.

Results and discussion

Searching chiral materials from cation-intercalated TMDs

Intercalated TMDs have recently attracted substantial interest due to their intriguing properties arising from intercalation applications^{49–51}. An important factor to be considered when intercalating a structure is that the host cannot be damaged significantly, such as a large-scale rearrangement of atoms or bonding distortions, which will remarkably increase the energy of the system and make the final structure unstable. The optimal solution to introduce chirality in layered TMDs could be adding screw rotation symmetry through the pivots within the vdW region, forming a chiral axis perpendicular to the vdW plane, which can guarantee minimal damage to the initial MX_2 motifs, e.g., a triangular prism in the 2H phase and an octahedron in the 1T phase. For simplicity, we consider cation intercalation in the $\sqrt{3} \times \sqrt{3} \times 2$ supercell of the 1T phase and the $\sqrt{3} \times \sqrt{3} \times 1$ supercell of the 2H phase with AA and AB stackings of MX_2 layers. There are two types of possible intercalation patterns in each supercell with intercalated polyhedra: (i) vertically or (ii) nonvertically distributed along the c axis (Figs. 1a, S1). Regarding the AB stacked 2H TMDs with type (ii) intercalation, there are two inequivalent configurations with the same space group (SG) $P6_3/22$ (belonging to the 65 Sohncke groups that preserve chirality)⁵² but opposite handedness in their intercalation patterns (see the circled red/blue spheres in Fig. 1a and the crystal structure in Fig. S1), in which the chiral symmetry is endowed by breaking the inversion and mirror symmetries of the pristine AB stacked 2H phase ($P6_3/mmc$). Regarding all the other intercalated situations, the products are achiral.

A series of new AB_3C_6 compounds can be obtained from the accessible TMD library⁵³ with intercalated TM cations by the requirement of valence state matching ($\chi^A + 3\chi^B = 6\chi^C$, assuming chalcogen with -2 common valence states) and inequivalent species on I(II)/III sites ($A \neq B$). All the 1T intercalated TMDs present CS symmetry, namely, $P\bar{3}1m$, $P\bar{3}1c$, $P6_3/m$, and $P6_3/mcm$. The 2H intercalated TMDs possess 1 CS ($P6_3/mcm$), 1 chiral ($P6_3/22$), and 2 non-CS ($P\bar{6}2m$ and $P\bar{6}2c$) structures (see Figs. 1b, S1). We evaluated the relative stability of the

intercalated TMD structures and noted that (i) the intercalated cations are in octahedral (triangular prism) motifs for AA stacked 1T and AB stacked 2H (AB stacked 1T and AA stacked 2H) phases; (ii) the intercalated octahedra tend to be nonvertically, rather than vertically, distributed along the c axis, as in the low-energy $P\bar{3}1c$ and $P6_322$ structures (Fig. S1). Such a nonvertical distribution of intercalated octahedra in the AA stacked 1T layers of pristine octahedra pertains to CS symmetry, whereas the nonvertical distribution of octahedra intercalated between the AB stacked 2H layers of triangular prisms leads to chiral symmetry ($P6_322$). We demonstrated the origin of how chirality is induced within the system (Figs. 1c, S2).

A number of chiral TMD materials ($P6_322$) are magnetic, including those that were synthesized in experiments, such as CoNb_3S_6 , FeNb_3S_6 , and MnTa_3S_6 ^{54–58}, and many others are nonmagnetic according to our DFT calculations (Fig. S3). We find a unique double hourglass electronic structure along the $\Gamma - \text{A}$ k -path around E_F for the nonmagnetic chiral TMDs (Fig. S4), as the unshelled d orbitals of their intercalated and/or host TM cations form partially occupied bands. In contrast, the double hourglass bands can be destroyed more or less in the magnetic case due to the broken time-reversal symmetry (T) (Fig. S5). Therefore, we focus our research on the nonmagnetic chiral TMD for studying the potential chirality-associated double hourglass fermion (Fig. 2a). Through careful examination of the electronic state and taking into account the material stability, we screened the best chiral TMD candidate, RhV_3S_6 ($P6_322$). The most stable Rh intercalated positions are I/II and I'/II' sites confirmed by globally detecting the total energy of the system when moving the cations in the vdW planes (Fig. S6). Further structural screening showed that RhV_3S_6 in other known AB_3C_6 structure types had higher energy than the $P6_322$ phase (Fig. 2b). The stability criteria in Fig. 2c, d show that RhV_3S_6 ($P6_322$) is dynamically stable (no occurrence of virtual frequencies in its phonon spectrum) and has a relatively large stability region (0.038 eV^2) with respect to its competing phases. Thus, all the stability criteria prove the stability of RhV_3S_6 , indicating the feasibility of experimental synthesis.

Double hourglass Weyl fermion in chiral TMD

The chiral TMD (RhV_3S_6) crystallizes in a hexagonal structure with SG D_6^6 ($P6_322$, No. 182), which presents semimetallic properties with valence and conduction bands crossing linearly at $(0, 0, \pm 0.447\pi)$ along $\Gamma - \text{A}$ without the SOC effect (Fig. 3a, b) and forms a fourfold degenerated hourglass Dirac semimetal state (Fig. S7 and Table S2). In the presence of SOC, the energy bands are generally nondegenerate, except at the time-reversal invariant

momenta (TRIM), e.g., Γ and A, and the hourglass bands split into two nested hourglass bands (Fig. 3c, d). Here, we provide a symmetry argument for the hourglass dispersions along $\Gamma - \text{A}$. The chiral TMD (RhV_3S_6) belongs to the nonsymmorphic space group $P6_322$, which contains a screw rotation operator

$$\tilde{C}_{2z}(x, y, z) \rightarrow \left(-x, -y, z + \frac{1}{2}\right) \quad (2)$$

where the tilde denotes a nonsymmorphic operation. The intercalated cations lead to chirality in the chiral TMDs ($P6_322$), whereas the nonsymmorphic \tilde{C}_{2z} symmetry is inherited from the parent 2H phase ($P6_3/\text{mmc}$), leading to hourglass Weyl fermions. In addition, T symmetry is preserved for nonmagnetic RhV_3S_6 . The T symmetry guarantees Kramers degeneracies at TRIM Γ and A due to $T^2 = -1$ in the presence of SOC. In the \tilde{C}_{2z} invariant k -path along $\Gamma - \text{A}$, we can choose each Bloch state $|u\rangle$ to be the eigenstate of \tilde{C}_{2z} . It is easily obtained that

$$\tilde{C}_{2z}^2|u\rangle = e^{-ik_z}|u\rangle, \quad (3)$$

which indicates that e^{-ik_z} is the eigenvalue of \tilde{C}_{2z}^2 . Thus, \tilde{C}_{2z} acts on the real-space part of the wavefunction and commutes with the time-reversal operator. Along the $\Gamma - \text{A}$ k line, each k point is invariant under \tilde{C}_{2z} , and the related Bloch state $|u\rangle$ can be chosen as the eigenstates of \tilde{C}_{2z} , namely, $\tilde{C}_{2z}|u\rangle = g_z|u\rangle$. One has $\tilde{C}_{2z}^2 = e^{-ik_z/2}$, and the eigenvalue of \tilde{C}_{2z} will be $g_z = \pm e^{-ik_z/2}$, i.e., $g_z = \pm 1$ at the Γ point $(0, 0, 0)$ and $g_z = \pm i$ at the point $(0, 0, \pi)$. Since \tilde{C}_{2z} always commutes with the time-reversal symmetry $T = K$ (without SOC) or $T = i\sigma_y K$ (with SOC), where K is the complex conjugate operator, the two Kramers pair $|u\rangle$ and $T|u\rangle$ must degenerate at the time-reversal invariant points Γ and A. In this case, the Kramers pair $|u\rangle$ and $T|u\rangle$ at the Γ point has the same eigenvalue, while the Kramers pair $|u\rangle$ and $T|u\rangle$ at the A point have the opposite eigenvalue. Such partner switching between Γ and A enforces a band crossing with twofold degeneracy along $\Gamma - \text{A}$. We further evaluated the double-valued representation of the double point group for each band and evidenced the state-switching characteristics, as seen from the degenerating and separating process of irreducible representations (irreps) at arbitrary point Δ along the $\Gamma - \text{A}$ line for both chiral S and R phases in Fig. 3e and Table S3. Here, Δ_i is the irrep obtained by scanning each hourglass band along the $\Gamma - \text{A}$ line, forming a degenerated partner with another Δ_j at TRIM. The partners Δ_{11} and Δ_8 (Δ_{10} and Δ_9) form the top left (bottom) corner of one hourglass band at Γ , separate from each other while leaving from the Γ point and recombine at the A point by exchanging the degenerated pair and constituting the right corners (Fig. S8). A similar

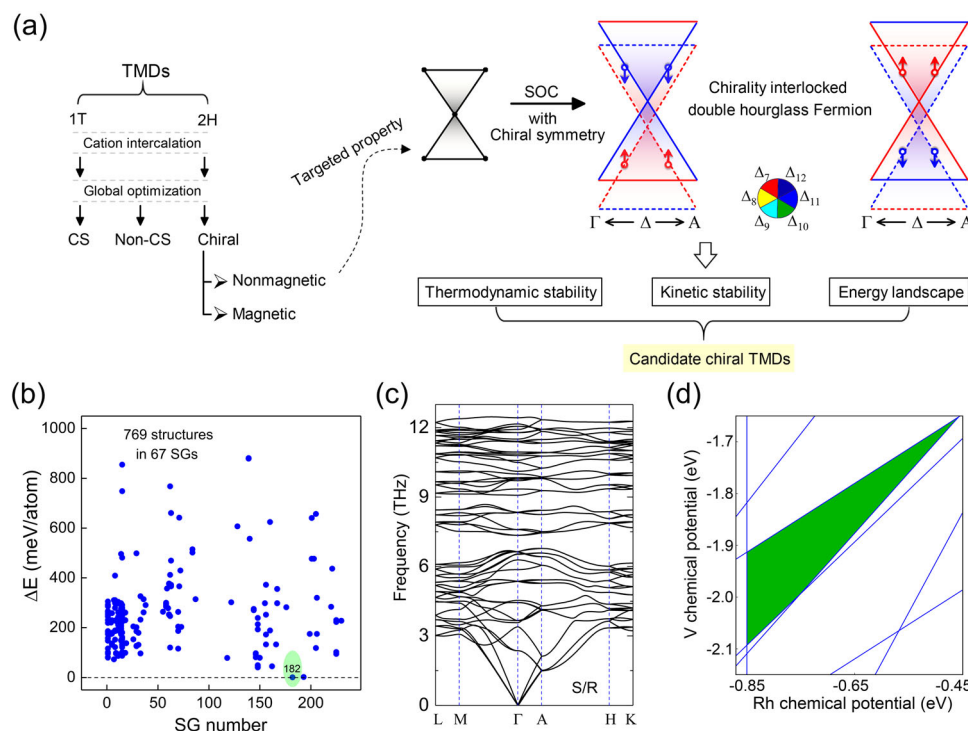


Fig. 2 Screening process and stability evaluation for stable chiral TMDs. **a** Design framework for the search of chiral TMDs with intercalated TM cations, targeting properties including explicit double hourglass fermions characterized by spin flipping between chiral structures with opposite handedness and structural stability. The potential chiral TMDs are divided into nonmagnetic and magnetic structures based on total energy evaluation. Subsequent electronic characteristic screening is performed for nonmagnetic structures with time-reversal symmetry. The stability of the screened chiral TMD was further evaluated by systematic material stability calculations. **b** Structural screening of RhV_3S_6 by comparison with all possible phases possessing the same stoichiometry (A:B:C = 1:3:6) distributed in 67 SGs. **c** Kinetic stability and **(d)** thermodynamic judgment for RhV_3S_6 . The nonimaginary frequency phonon dispersion suggests the most stable ground state for the chiral phase.

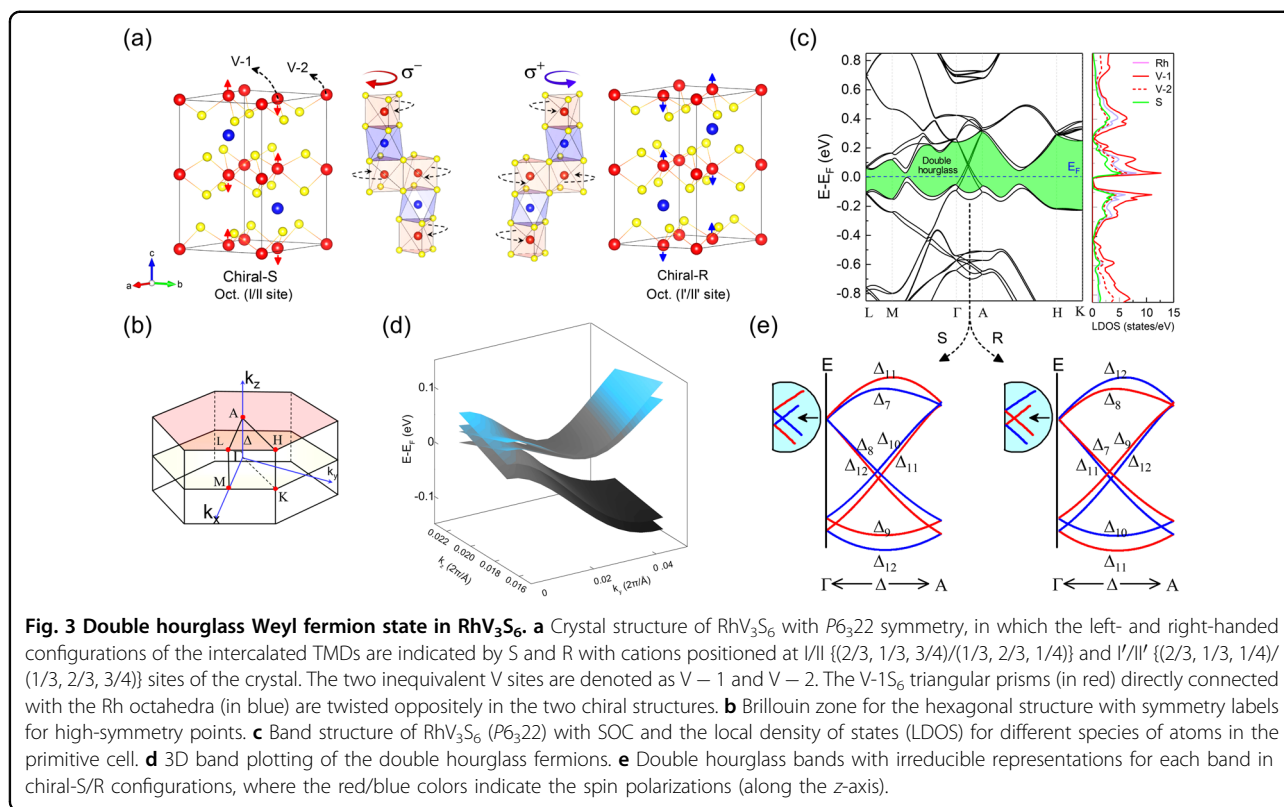
situation also holds for the other nested hourglass band formed by Δ_7 and Δ_{12} (Δ_{11} and Δ_{12}) degenerated partners. As a result, the occurrence of partner switching while moving from one TRIM to the other on the $\Gamma - A$ line leads to the nontrivial band connectivity diagram and the nested double hourglass bands in RhV_3S_6 . Furthermore, we note that the spin characteristics for every hourglass band are completely opposite between chiral-S/R structures for the chiral TMDs, which indicates the interlocking of spin-momentum of the double hourglass bands and crystalline chirality,

$$\psi(r, \uparrow) \xrightarrow{S/R} \psi(r, \downarrow). \quad (4)$$

Here, the r stands for spatial coordinates. It should be noted that the linear energy dispersion of the double hourglass bands extends over a relatively large energy range and thus could be experimentally observed uncomplicatedly *via* various spectroscopy techniques (e.g., tunneling spectroscopy, photoemission, or transport experiments)^{32,59}.

Tuning the dia-chiral response and double hourglass state by strain

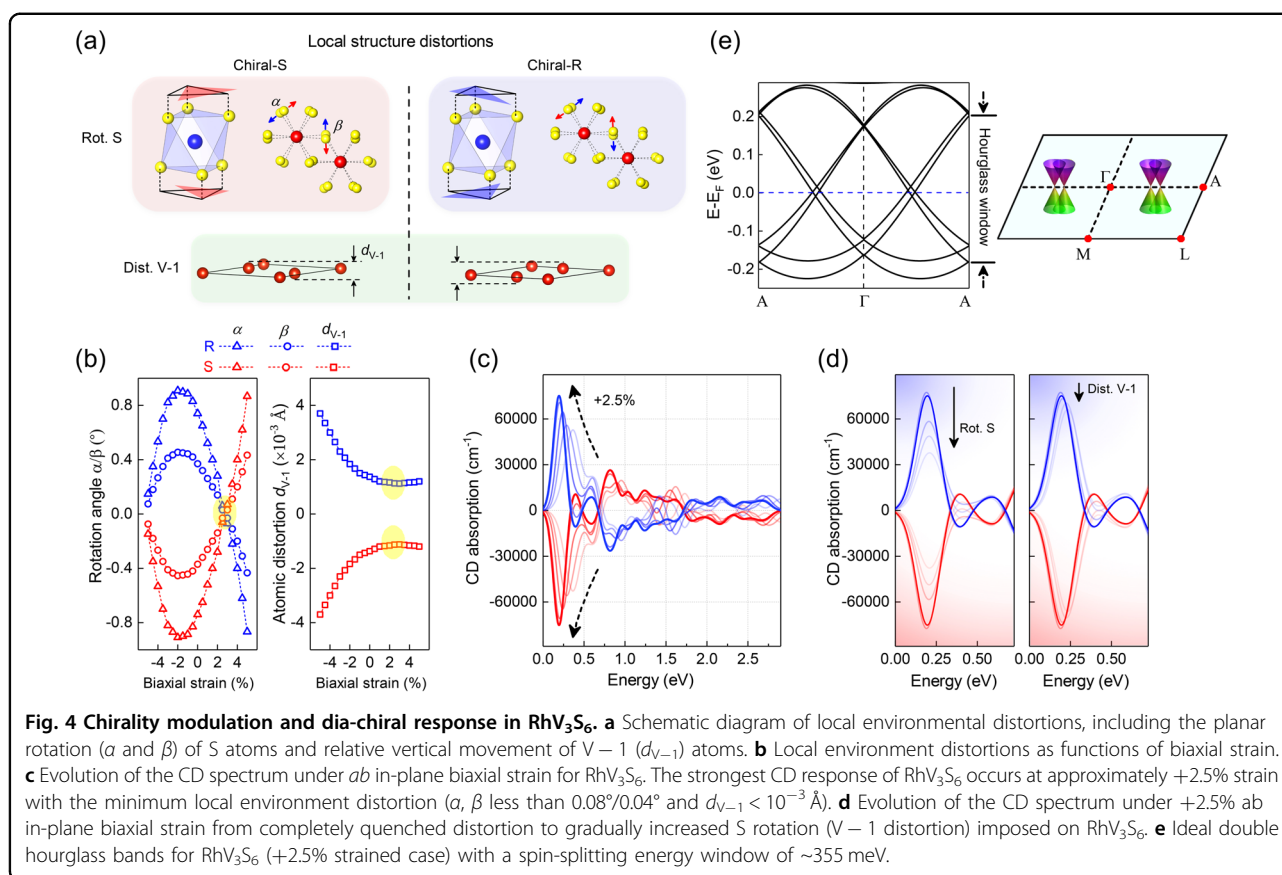
The interlocking of crystalline chirality and double hourglass bands indicates the possibility of tuning one through another. The left-/right-handed RhV_3S_6 configurations are determined by the placement of Rh octahedra (at I/II and I'/II' sites) and present opposite CD responses (Fig. 4). The octahedron in pristine RhV_3S_6 maintains a slight atomic distortion for the V – 1 and S atoms: the V – 1 atoms that are originally located on the ab plane have relative displacements along the c axis ($d_{V-1} = 0.0015 \text{ \AA}$); the S atoms rotate in the ab plane ($0.594^\circ/0.297^\circ$ for α and β , respectively). In general, the local atomic distribution and bonding environment could affect the chirality of the system. The enhanced distorting operation (e.g., rotation) is usually considered the origin of chirality or the main contributor^{60–62}. However, not all atomic bonding distortions contribute to the chirality, such as the Kekulé distortion⁶³ for epitaxial graphene on a copper substrate that breaks the chiral symmetry⁶⁴. Since distortion can increase or decrease chirality, one fundamental question is whether an intrinsic chirality hidden in



a specific microstructure exists and whether local atomic distortion will force it to decay. Here, we probed this issue in our predicted chiral TMD – RhV_3S_6 , as the inherent chirality is intimated with the special configuration of the intercalated octahedra (Fig. 1c) and can be sensitively captured by the double hourglass fermion by controlling the intercalation-induced local structure distortions. This is done by applying biaxial strains (ab in-plane) on RhV_3S_6 . We note that as the perturbation is induced *via* strain, the relative movement of V – 1 diminishes gradually, whereas the rotation angle of S experiences a certain degree of fluctuation (Fig. 4b). Both of these structural deformation factors reach the critical point around the +2.5% strained case, as the octahedron recovers to its pristine structure with barely any rotation. Additionally, the CD intensity presents the maximum for the pristine octahedron stacking configuration, while both the enhanced perturbation of S (α and β) and V – 1 (d_{V-1}) distortions weaken the CD signal (Fig. 4c). Even for the slightly distorted octahedra in unstrained RhV_3S_6 , the CD response is weakened. To further demonstrate the intrinsic chirality restored within intercalated octahedra, we separately evaluated the CD response under +2.5% strain with increasing S (Rot. S; α, β) and V – 1 (d_{V-1}) distortions, starting from the structure with quenched atomic distortion, which indicates the compensation of intrinsic chirality (for quenched distortion,

see the curves with the highest CD peaks in Fig. 4d). Indeed, the intrinsic chirality of RhV_3S_6 is introduced by the pristine octahedron placement configuration, forming the intrinsic chiral domain of the crystal. However, the domain steady state will be broken by structural perturbation due to redistributed state coupling (i.e., rebalancing of the wavefunction), leading to the opposite handedness and weakening of the intrinsic chirality, as an inevitable “dia-chiral response” that can be modulated by external strain, as discussed above.

In the “dia-chiral response” modulation, we find that the double hourglass state presents a tunable hourglass window, namely, the covered spin-splitting energy range in E - k space by hourglass bands. The splitting window for pristine RhV_3S_6 reaches a record energy range of ~380 meV (Fig. 3c), which is significantly larger than those in KHgSb (60/80 meV for $\tilde{Z}/\tilde{X} - \tilde{\Gamma}/\tilde{U}$ at 010-surface)²², Ag_2BiO_3 (3 meV for $X - S$ k line)²⁷, and Bi/Cl-SiC with a 111-plane (150/187 meV for $X/\Gamma - M/Y$ direction). Strain engineering enables the wide-range modulation of the double hourglass bands (~300–440 meV) for RhV_3S_6 (Fig. S9). In the +2.5% strained case with the maximum CD intensity, the middle band-crossing points (the normal centers of the double hourglass bands; analogous to the Weyl points in achiral TMD Weyl semimetals^{13–17}) shift to the E_F (Fig. 4e). In fact, the double hourglass bands are dominated by V 3d



orbitals hybridized with Rh 4*d* and S 3*p* components, forming anti-bonding states (Figs. 3c, S10, S11). When *ab* in-plane tensile strain is applied, the heights ($h_{\text{Oct.}}$ and $h_{\text{Tri.}}$) of the octahedron and triangular prism are both compressed (Fig. S11), and the Rh-S and V-S bond lengths (d_{RhS} and d_{VS}) are enlarged. As a consequence, the *p-d* coupling between the V 3*d* (or Rh 4*d*) and S 3*p* orbitals is weakened due to the decreased overlap between their wavefunctions, which tends to lower the anti-bonding double hourglass states. Therefore, the double hourglass bands continuously move down with a decreasing splitting window, presenting flexible tunability along with the modulation of crystalline chirality.

Conclusions

In summary, we predict a number of octahedral intercalated chiral TMDs with a type of spin-split double hourglass Weyl fermion, which is interlocked with crystalline chirality. The optical CD of intercalated TMDs is evaluated to monitor the chirality of the material that can be tuned by external strain. The best candidate RhV₃S₆ is predicted to be thermodynamically stable, whose synthesis is called for as the first example of a chirality-enforced double hourglass fermion. On the other hand, this type of

chiral TMD may provide a new platform for exploring tunable chiraltronics within a three-dimensional scale, e.g., a chiral plasmonic device^{65–67}, as three-dimensional chiral plasmonic structures that exhibit a strong intrinsic chiral optical response remain an extreme challenge. Furthermore, the predicted AB₃C₆ family members could also raise great interest in extraordinary phenomena and device applications, such as the anomalous Hall effect⁵⁴, mesoscopic magnetic modulation⁶⁸, three-state nematicity⁶⁹, and tunable giant exchange bias by spin glass⁷⁰. The stable properties and facile synthesis of these chiral TMDs guarantee the subsequent micro/nanoscale process and device integration. Our study provides an in-depth mechanism of intrinsic chirality induction through polyhedral intercalation in TMDs, which have a significant chiral optical response. More importantly, insight into introducing intrinsic chirality through chiral intercalation of polyhedra in achiral systems to realize a strong chiral response will shed light on designing/searching for new chiral structures extensively with novel characteristics. Our findings are one step forward in the exploration of chirality-related hourglass Weyl fermions in inorganic compounds and provide a new understanding of structural chirality for future chiral-functionalized optoelectronic devices.

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Author contributions

X.Z. directed the research project. P.H. performed the calculations and prepared the figures. P.H., Q.L., R.W., H.S., and X.Z. conducted the symmetry analysis of the chiral TMDs. X.C., X.Z., and H.Z. evaluated the stability of these compounds. P.Z., S.X., and W.X. analyzed the state coupling and spin characteristics of RhV₃S₆. All authors contributed to the discussion and provided inputs to the paper. P.H., X.C., and P.Z. contributed equally to this work.

Competing interests

The authors declare no competing interests.

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