Supplementary Information

Uncovering and tailoring hidden Rashba spin-orbit splitting in

Supplementary Note 1: Identification of the two inversion-partner sectors in ferroelectric SnTe and illustration of how the R-1 spin-splitting originates from the local symmetries of the sectors rather than being a consequence of the global inversion symmetry breaking.

At room temperature, !-SnTe has the centrosymmetric rocksalt (space group Fm-3m) structure¹ (Supplementary Figure 1a) and, as temperature is lowered,² it undergoes a (ferroelectric) phase transition to the non-centrosymmetric rhombohedral (space group R3m) R-1 structure,^{3, 4} where the Te atom is spontaneously displaced along the [111] direction relative to Sn atom. We focus on the bands near Fermi energy around the Z point which exhibits the most pronounced Rashba spin splitting (Supplementary Figure 1b).

In the centrosymmetric rocksalt phase (illustrated by dashed lines in

Supplementary Figure 1. Non-centrosymmetric rhombohedral -SnTe having strong R-1 effect. a, Crystal structure of centrosymmetric !-SnTe identify with two inversion sectors (pink domain) and (cyan domain). The centrosymmetric phase transfers to a non-centrosymmetric rhombohedral phase as the Te atom displaced from Sn clong [111] direction b. Band structure around the time reversal invariant

the Te atom displaced from Sn along [111] direction b, Band structure around the time-reversal invariant momentum

Non-symmorphic screw axis symmetry enforces the four-fold

degeneracy of the

The rotation symmetry of these eight symmetry operators forms the little group D_{2h} of the --point. The character table of D_{2h} is given in Supplementary Table 1. Note that $BaNiS_2$ as a non-magnetic crystal also contains the time reversal symmetry $\hat{}$.

(1) The effective model Hamiltonian in the absence of SOC

The distinct band topology along - and - of the monolayer BaNiS₂ without SOC shown in Figure 1c can be understood from the effective model Hamiltonian. Since the Rashba bands of our interesting around the --point are mainly derived from the Ni *d*-orbits with and character, the atomic *d* states of Ni atom , where *d* denotes a hybridization state of and , are the reasonable choice as the basis of the invariant expansion. The Bloch basis \rangle can be explicitly expressed as a Bloch sum of the local *d* states,

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direction, obtaining forbidden interaction between degenerate states segregated on two inversion-partner sectors and leading to the two-fold degeneracy of the energy bands in agreement with the first-principles calculation,