Higher-dimensional spin selectivity in chiral crystals

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The chiral-induced spin selectivity effect has attracted significant attention in interdisciplinary research fields due to its potential to offer pure spin currents without requiring magnetic fields or materials. Previous studies have neglected the spin selectivity in the normal plane of chiral molecules and crystals because the chiral-induced spin-orbit coupling (CISOC) strength is assumed to be substantially stronger than the antisymmetric SOC (ASOC) terms. This poses a challenge in chiral van der Waals crystals like indium selenoiodide (InSeI) that consists of heavy elements in a bundle of atomically precise 1D helical chains. This study aims to investigate the interplay between CISOC along the screw axis and ASOC in the normal plane within a chiral crystal, using both general model analysis and first-principles simulations. Using InSeI as a showcase, we reveal a three-dimensional (3D) spin selectivity in chiral crystals. The resulting phase diagram of spin accumulation shows the potential for controlling phase transition and flipping spin by reducing symmetry through surface cleavage, thickness reduction, or strain. We also experimentally synthesized high-quality InSeI crystals of the thermodynamically stable achiral analogue, which showed exposed (110) facets corresponding to single-handed helices to demonstrate the potential of material realization for higher-dimensional spin selectivity in the development of spintronic devices.