To the Editors of Nature Materials,

There has been great progress in the past few years in the field of topological electronic structure, starting with the theoretical understanding of quantum spin Hall systems and topological insulators, followed by the proposal of specific realizations of these phases, and finally the experimental verification of these findings. This remarkable success stands in contrast to the limited progress towards a realization of the related topological phase known as a Chern insulator, or quantum anomalous Hall insulator, despite its being proposed theoretically over two decades ago. In this work, we propose a new search strategy for achieving an experimental realization of a Chern insulator which avoids many of materials-related difficulties of previous proposals, we verify its viability with first principles calculations, and we discuss the possibilities for an experimental realization.

Previous proposals for making a Chern insulator largely depend on doping a Dirac cone material (e.g. graphene,  $Bi_2Se_3$ , HgTe quantum wells) with magnetic impurities to break time reversal symmetry. These proposals face a variety of materials challenges related to controlled doping of small-band-gap materials and alignment of the spins of the disordered dopants. Our proposal is to deposit submonolayer coverages of heavy atoms with large spin-orbit coupling (e.g. Bi) onto the surface of a large-band-gap magnetic insulator. We argue that this combination of materials has the appropriate combination of magnetic exchange, spin-orbit coupling, and surface dispersion to generically produce band structures with isolated surface bands displaying non-zero Chern numbers. These properties combine to produce materials with anomalous Hall coupling on the order of  $e^2/h$ , and finding a Chern insulator then reduces to searching for examples with a global energy gap. In addition, by starting with large-gap materials with naturally aligned spins, we avoid many of the materials difficulties of previous work.

In our paper, entitled "Chern insulators from heavy atoms on magnetic substrates," we present our search strategy and furthermore use first-principles calculations to prove its viability. Our calculations reveal examples of Chern insulators with band gaps as large as 140 meV, to our knowledge the largest gap of any proposed Chern insulator. In addition, we discuss the possibilities for a combined experimental/theoretical effort to realize these ideas. Our paper thus meets the criteria of significance and interest to both experimental and theoretical readers and warrants publication in Nature Materials.

Thank you for your consideration,

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