

## Chern insulator at a magnetic rocksalt interface

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Considerable efforts have recently been devoted to the experimental realization of a two-dimensional Chern insulator, i.e., a system displaying a quantum anomalous Hall effect. However, existing approaches such as those based on magnetic doping of topological-insulator thin films have resulted in small band gaps, restricting the effect to low temperatures. We use first-principles calculations to demonstrate that an interface between thin films of the topologically trivial ferromagnetic insulators EuO and GdN can result in a band inversion and a nonzero Chern number. Both materials are stoichiometric and the interface is nonpolar and lattice-matched, which should allow this interface to be achievable experimentally. We show that the band structure can be tuned by layer thickness or epitaxial strain, and can result in Chern insulators with gaps of over 0.1 eV.

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Recently, there has been considerable interest in experimental attempts to realize two-dimensional Chern insulators [1–4], also known as a quantum anomalous Hall insulators. A Chern insulator, which by definition is an insulator whose occupied bands carry a nonzero Chern number [5], is a species of topological insulator with broken time reversal (TR) symmetry. Such a material displays the essential physics of the integer quantum Hall effect, including robust dissipationless quantized conducting edge states, but without an external magnetic field. These edge states could be used in electronics applications, and Chern insulators would display strong magnetoelectric effects [6].

While the requirements for a Chern insulator, namely broken TR symmetry and spin-orbit coupling (SOC), are in principle very common, finding a robust experimental realization of a Chern insulator has proven very challenging. The first successful demonstration consisted of doping a two-dimensional slab of the  $Z_2$  topological insulator  $(\text{Bi,Sb})_2\text{Te}_3$  with Cr atoms [1]. The Cr dopants order ferromagnetically at low temperatures, breaking TR symmetry and driving the system into a Chern-insulating state. Unfortunately, the magnetic ordering temperature of this system is only 15 K, and  $\sigma_{xy}$  becomes quantized to better than 10% only below about 400 mK, greatly limiting potential applications. Furthermore, careful control of the Bi and Sb concentrations is needed to separate the surface states from the bulk states, and gating is needed to adjust the Fermi level into the gap. Similar proposals that focus on magnetically doping 2D slabs of other topological insulators [7], quantum spin Hall insulators [8], or graphene [9] will also likely be limited to very low temperatures and small band gaps.

In this work, we focus on a particular realization of an alternate strategy for designing Chern insulators. This strategy, which consists of directly combining magnetic insulators with materials that have large SOC, has the advantage that we are not limited to known topologically nontrivial materials. As a result, we can choose to combine materials with large band gaps and robust magnetic orderings, potentially allowing for high-temperature operation. We have previously used this strategy to show that placing heavy adatoms on the surface of a magnetic insulator frequently produces topologically nontrivial band structures, some of which are insulating [10]. In this work, we take the complementary

strategy of combining two trivial magnetic insulators in such a way as to get a nontrivial band crossing at the interface between them, resulting in a Chern insulator.

We focus on the interface between two magnetic rocksalt materials, EuO and GdN. EuO is well known for being one of the few ferromagnetically ordered semiconductors, with a Curie temperature of 70 K and a band gap of 1.1 eV [11]. Similarly, GdN is ferromagnetically ordered with a Curie temperature of 58 K [12], and while it is a metal in the bulk [13], it is known to become insulating in thin-film form [14]. Experimentally, the lattice constant of EuO is 5.14 Å [15] and that of GdN is 4.99 Å [12], resulting in an experimentally reasonable 3.0% mismatch. We show that for a proper choice of strain and layer thickness, the interface between EuO and GdN can result in a Chern-insulating state with a band gap of over 0.1 eV. While the Curie temperatures of these materials still fall below room temperature, such a system would be a significant step in the direction of a robust room-temperature Chern insulator.

We perform first-principles density-functional theory (DFT) [16,17] calculations using the VASP code [18,19] and PAW potentials [20,21], with SOC included. We carry out structural relaxations using the PBE generalized-gradient approximation (GGA) [22], with a Hubbard  $U$  (DFT +  $U$ ) [23,24] correction of 6.5 eV on the Eu and Gd  $f$  states. Because the band alignments at the interface are of special importance in this work, we then perform a final band-structure calculation using a hybrid functional. We adopt the HSE functional [25], which is known to provide an accurate description of the bulk band structure and magnetic properties of both EuO and GdN [26–28]. This success of the HSE hybrid functional should not be surprising in view of the fact that  $\text{Eu}^{2+}$  and  $\text{Gd}^{3+}$  are both closed-shell  $f^7$  ions, so that correlation effects are mild. Results from our DFT calculations are then used as input to construct maximally localized Wannier functions using WANNIER90 [29,30]. Chern numbers and band gaps are calculated using Wannier interpolation of the band structure; the Chern numbers are computed by sampling the Brillouin zone (BZ) by a dense  $k$ -point grid and summing the Berry phases around the loops formed by each set of four adjacent  $k$  points.

We begin by outlining our basic strategy for constructing a Chern insulator. Our goal is to find two topologically trivial materials that, when placed together, result in a band

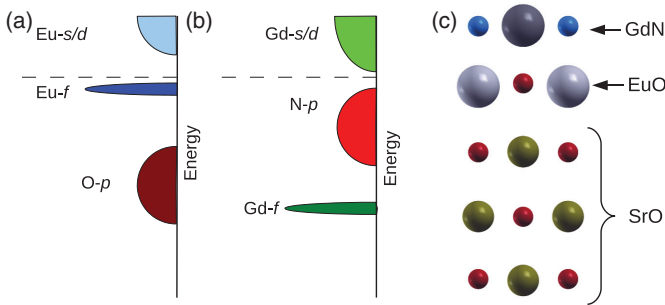


FIG. 1. (Color online) (a) Schematic density of states of EuO. (b) Same for GdN. (c) Side view of proposed interface structure, composed of one GdN layer atop one EuO layer on a SrO substrate. Large atoms are cations (Gd, Eu, and Sr); small atoms are anions (N and O).

inversion and a Chern insulator. The (001) surface of EuO is nearly unique among binary compounds in providing a simple insulating nonpolar surface that breaks time-reversal symmetry [31], making it a good starting point for our strategy. One drawback is that even bulk EuO has a Curie temperature of only 70 K, and measurements show reductions for thin films, e.g., to 30 K in 1 nm thick films of EuO [32–35] (comparable to our thicker examples). However, the bulk Curie temperature of EuO can be increased by pressure [36], strain [34,37], and doping [38], so further engineering of the transition temperature may be possible. We assume here that all spins point normal to the surface; if not, we propose that a small external magnetic field can be used to align the spins in this direction, in view of the small magnetocrystalline anisotropy of our cubic materials [39].

Both the valence and conduction bands of EuO consist of strongly spin-polarized bands localized on the Eu atom, as shown schematically in Fig. 1(a). Eu ( $Z = 63$ ) provides strong SOC, making EuO an excellent candidate material. In fact, our calculations show that a single layer of EuO will become a Chern insulator under sufficiently strong compressive epitaxial strain, which reduces the band gap and eventually causes the conduction band to overlap with the valence band. Unfortunately, the strain necessary to cause this band inversion is unrealistically large ( $\sim 10\%$ ), forcing us to look for a second material to combine with EuO to achieve the same effect under more realistic conditions. This second material should provide conduction bands of the correct symmetry such that they result in an avoided crossing and a transfer of Chern number when they overlap with the occupied Eu  $f$  states.

There are many candidate materials with the rocksalt structure that may be possible to interface epitaxially with EuO, but the majority of them are highly ionic materials with very large band gaps, making them unsuitable for this application. One exception is CdO, which has been studied previously in quantum wells and superlattices with EuO as a candidate topological system [40]. In agreement with previous results, we find that this combination does produce the desired Chern-insulating state for certain values of layer thickness and strain, but with a small band gap. The small gap results from the weak interaction between the Cd  $s$  and Eu  $f$  states near  $\Gamma$ ,

which can be understood in the context of the Wigner-Eckhart theorem; i.e., a first-order  $\mathbf{k} \cdot \mathbf{p}$  perturbation ( $\Delta l = 1$ ) cannot link  $l = 0$  and  $l = 3$  states, at least without assistance from the cubic crystal field ( $\Delta l = 2$ ). In addition, the Cd  $s$  states have only a weak spin splitting arising from exchange coupling to the Eu; this results in a very limited phase space for nontrivial topological behavior before the Cd  $s$  state of opposite spin crosses the Fermi level and the system becomes metallic. Finally, CdO is poorly lattice-matched with EuO, which would make synthesis of these structures challenging.

Based on the example of CdO as well as general considerations of experimental feasibility, we conclude that the ideal rocksalt material to pair with EuO would have the following characteristics: (1) a similar lattice constant to EuO, (2) a large SOC, (3) a small band gap, (4) spin-polarized conduction bands with  $d$  character, and (5) a conduction band minimum at  $\Gamma$  in the surface BZ. The only material we know of that meets all of these requirements is GdN, a ferromagnetic spin-polarized semimetal in bulk which becomes insulating in thin-film form.

In Figs. 1(a) and 1(b) we present a schematic band-alignment diagram for GdN and EuO. We seek to engineer a band inversion between the Eu  $f$  state at the top of the valence band of EuO and the Gd  $d_{x^2-y^2}$  orbital at the bottom of the conduction band of GdN by creating an interface between these two materials. In order to obtain the largest possible coupling between the two materials, we begin with single layers of EuO and GdN stacked on an SrO substrate, as shown in Fig. 1(c). We show the resulting surface band structure in Fig. 2, with all spins aligned ferromagnetically perpendicular to the surface, for two different values of in-plane lattice

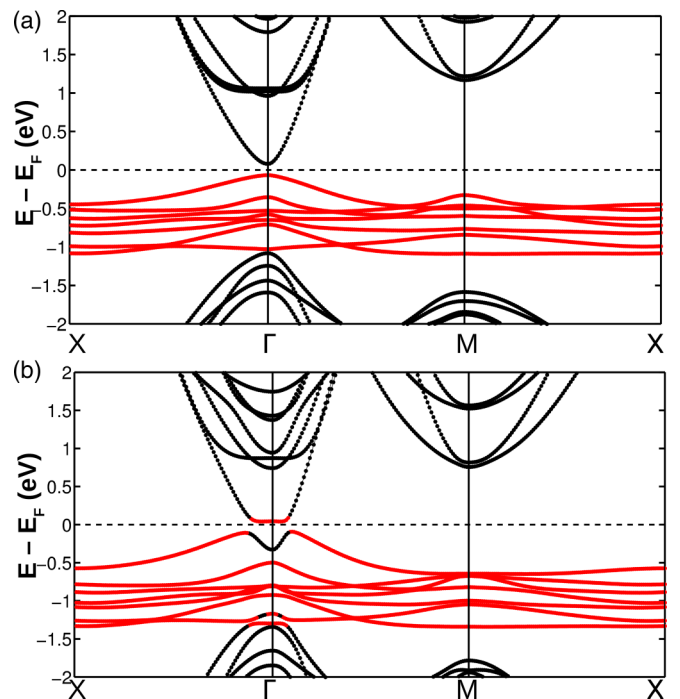


FIG. 2. (Color online) Band structures of one GdN layer atop one EuO layer on an SrO substrate at (a) 3.62 Å ( $C = 0$ ), and (b) 3.44 Å ( $C = -1$ ). Majority Eu  $f$  states are plotted in red; others are in black.

TABLE I. Computed Chern number  $C$  and band gap  $E_g$  for  $N_{\text{GdN}}$  layers of GdN atop  $N_{\text{EuO}}$  layers of EuO on an SrO substrate of the specified lattice constant  $a$ .

$N_{\text{EuO}}$	$N_{\text{GdN}}$	$a$ (Å)	$C$	$E_g$ (meV)
1	1	3.62	0	111
1	1	3.53	-1	3
1	1	3.48	-1	130
2	1	3.62	-1	80
2	1	3.53	-1	123
1	2	3.48	-1	71
2	2	3.62	-1	62

constant. At large lattice constants this interface is a trivial insulator, as shown in Fig. 2(a). However, we find that the band gap at  $\Gamma$  is sensitive to epitaxial strain, and decreases by  $\sim 50$  meV for every 1% compressive strain, as shown in Table I. This dependence of the gap on strain carries over to the thicker slabs investigated later in this work, and makes the band structure of this system widely tunable. Reducing the lattice constant of this system to the critical value of 3.53 Å causes the Gd  $d_{x^2-y^2}$  conduction-band minimum to cross the Eu  $f_{x^3-iy^3}$  valence-band maximum at  $\Gamma$ . Further reduction of the lattice constant, as shown in Fig. 2(b), results in an avoided crossing and a transfer of Chern number from the valence band to the conduction band, leaving the occupied bands with a Chern number of  $-1$ .

By varying the strain in the system, the band gap can be increased above 0.1 eV, as shown in the first three rows of Table I. This very robust gap can be traced to several factors. First, the atomically thin layers of GdN and EuO confine the relevant states at the interface, resulting in a large overlap. Second, as discussed above, the  $f$  and  $d$  character of the bands allows for first-order coupling between them. Finally, the large separation of the conduction-band minimum and valence-band maximum from other bands in the system allows for a large band inversion and a strong avoided crossing. This isolation of the relevant bands also allows for a relatively wide range of strains that can result in a Chern-insulating state, which should make our predictions more robust and easier to achieve experimentally.

The value of the Chern number in this system can be understood by examining the symmetries of the bands that take part in the avoided crossing [41]. At the  $\Gamma$  point of the BZ, the bands all belong to one of four nondegenerate irreducible representations that can be labeled by the eigenvalues of the fourfold rotation operator. In the case of a single band crossing where the eigenvalues of the bands differ only by a factor of  $\pm i$ , the exchange of Chern numbers is uniquely determined [42]. In the case of GdN on EuO, where the conduction-band minimum has the symmetry of a  $d_{x^2-y^2}$  orbital, these symmetry considerations imply that  $C = -1$ , consistent with our direct numerical calculations. In the case of CdO, the conduction band minimum has  $s$  character, and  $C = 1$ .

Because the topological behavior of this system is determined by the symmetries of the bands involved in the crossing, our results should be robust against many of the difficulties normally associated with first-principles calculations of materials with rare-earth elements. These band symmetries are

determined by the bulk band structures, which are known experimentally and are well described by our hybrid-functional calculations, and the electrostatic environment of the film surface termination. While the exact alignment of the Gd  $d$  and Eu  $f$  bands is difficult to calculate precisely, we have shown that this alignment is tunable, lending confidence to our prediction of a Chern-insulating state somewhere in the parameter space. Finally, we note both Eu(2+) and Gd(3+) have closed  $f$  shells, and Gd in particular strongly prefers a (3+) valence, which limits the possibilities for valence fluctuations and strong correlation effects in this system.

In order to gain some insight into the behavior of this system, we build a simple two-band single-spin tight-binding model, in the spirit of the Haldane model [43], but here designed to capture the key behavior of the valence-band maximum and conduction-band minimum of our system. The model, which treats only the majority spin channel in the ferromagnetic phase, consists of a square lattice with a single  $d_{x^2-y^2}$  orbital at  $(0,0)$  and a single  $f_{x^3-iy^3}$  orbital at  $(\frac{1}{2}, \frac{1}{2})$ , labeled by  $a = \{1, 2\}$ . These orbitals have on-site energies  $\pm \Delta$ , and strong hoppings ( $t_1 < 0, t_2 > 0$ ) to nearest neighbors of the same sublattice. The sublattices are coupled by a weaker complex interaction term,  $\lambda_{ij} = u \hat{e}_{ij} \cdot (\hat{x} - i \hat{y})$ , where  $u$  is the magnitude of the coupling and  $\hat{e}_{ij}$  is the direction of the hopping. This results in the Hamiltonian

$$H = \sum_{i,a} (-1)^{a+1} \Delta c_{ia}^\dagger c_{ia} + \sum_{\langle ij \rangle a} t_a c_{ia}^\dagger c_{ja} + \sum_{\langle ij \rangle} \lambda_{ij} c_{i1}^\dagger c_{j2} + \text{H.c.} \quad (1)$$

The sums over  $i$  and  $j$  are over unit cells, and the sum over  $a$  is over the two orbitals. Values of  $t_1 = -1.5$  eV,  $t_2 = 0.15$  eV, and  $u = 0.15$  eV provide a qualitative fit of the band structure, and the effect of strain can be mimicked by varying  $\Delta$  from 3.15 eV to 3.35 eV, corresponding to  $a = 3.44$  Å and  $a = 3.62$  Å, respectively (see Fig. 2). The Chern-insulating state at small values of  $\Delta$  is separated from the trivial state at large  $\Delta$  by a band touching at  $\Gamma$  at the critical value of  $\Delta_c = -2t_1 + 2t_2$ . For  $\Delta$  slightly below  $\Delta_c$ , the band gap of the Chern-insulating phase increases linearly with  $\Delta_c - \Delta$ , but for larger band inversions, the band gap of the Chern-insulating state begins to saturate, growing like  $(\Delta_c - \Delta)^{\frac{1}{2}}$  as the avoided crossings move away from  $\Gamma$ . This behavior is consistent with our first-principles results showing increasing gaps for larger band inversions, as long as the two orbitals near the Fermi level remain separated from other bands in the system.

Returning to the first-principles results, we next consider thicker layers of both GdN and EuO. These thicker layers are likely to be easier to grow and measure experimentally, as well as more likely to have bulklike magnetic ordering temperatures. First, we consider an additional GdN layer, as shown in Fig. 3(a). Consistent with our expectations, an additional layer of GdN results in a lower conduction-band minimum at a given lattice constant, consistent with the fact that thicker slabs of GdN should approach bulklike semimetallic behavior. This behavior is convenient because it allows the band structure to be tuned not only by strain, but also by varying the GdN layer thickness. We expect that as the GdN thickness increases, the relevant conduction-band state

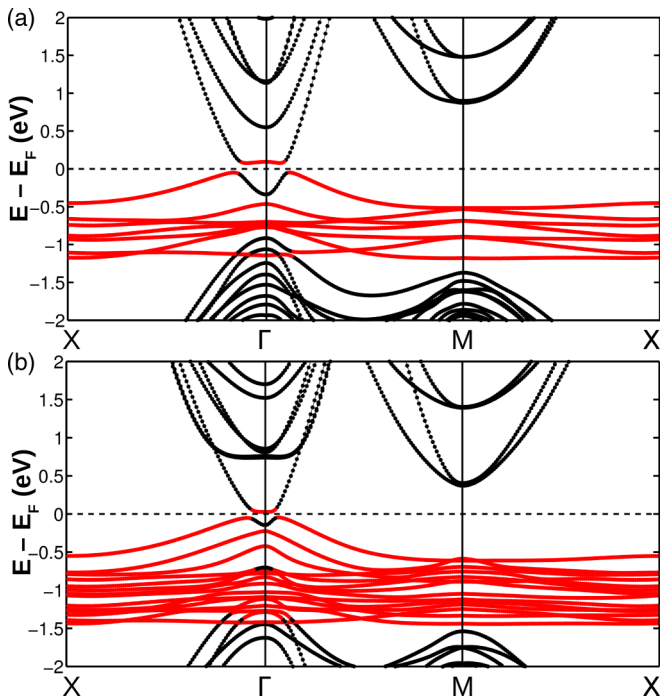


FIG. 3. (Color online) Band structures of (a) two GdN layers atop one EuO layer on SrO at 3.53 Å, and (b) one GdN layer atop two EuO layers on SrO at 3.48 Å. Both have  $C = -1$ . Colors as in Fig. 2.

will become more delocalized, leading to less interaction with EuO and a smaller band gap. However, this effect appears to be fairly weak when going from one to two layers, as shown in Table I.

We also explore the band structure of a single GdN layer on top of two EuO layers, as shown in Fig. 3(b). This configuration also results in a Chern insulator for appropriate values of strain, but the additional Eu  $f$  levels from the second EuO layer limit the total band gap. This occurs because of the relatively weak

splitting between the  $f$  orbitals on the two Eu atoms; for large band inversions, the  $d_{x^2-y^2}$  band from the Gd crosses several of the Eu- $f$  states, closing the band gap. However, as shown in Table I, the splitting between different Eu  $f$  levels is still large enough to allow significant band gaps, even for thicker EuO layers.

In all of the above calculations, we have assumed that the spins in both EuO and GdN align ferromagnetically, as they do in the bulk. As a preliminary test of this hypothesis, we consider an antiferromagnetic arrangement with a single spin-up GdN layer atop a single spin-down EuO layer on SrO at 3.53 Å, and we find that the antiferromagnetic configuration is 0.5 meV higher in energy than the ferromagnetic configuration considered previously. This energy difference is consistent, in sign and magnitude, with the calculated exchange parameters in bulk GdN and EuO [26,27]. Further calculations would be needed to determine all of the relevant magnetic exchange parameters, as well as the effects of reduced dimensionality on the magnetic ordering temperature. Also, as mentioned earlier, we have assumed that the spins are aligned along  $z$ , which could require the application of a small external field, as the spins lie along the (111) direction in bulk EuO [39]. Nevertheless, with these qualifications, we believe the needed magnetic structures should be attainable experimentally.

To summarize, our proposal results from a design strategy in which known topologically trivial materials with strong spin-orbit coupling are combined in such a way as to engineer a band inversion, resulting in robust, topologically nontrivial behavior. We have shown that the (001) interface between GdN and EuO is an excellent candidate system for achieving a robust Chern-insulating state at temperatures of up to 70 K and with band gaps of over 0.1 eV. This nonpolar lattice-matched interface consists of known stoichiometric magnetic insulators, which should make it achievable experimentally.

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