This is the accepted manuscript made available via CHORUS. The article has been published as:

**GdN thin film: Chern insulating state on square lattice**  
Zhi Li, Jinwoong Kim, Nicholas Kioussis, Shu-Yu Ning, Haibin Su, Toshiaki Iitaka, Takami Tohyama, Xinyu Yang, and Jiu-Xing Zhang  
Phys. Rev. B 92, 201303 — Published 10 November 2015  
DOI: [10.1103/PhysRevB.92.201303](https://doi.org/10.1103/PhysRevB.92.201303)
GdN thin-film: Chern Insulating State on Square Lattice

Zhi Li,† Jinwoong Kim,‡ Nicholas Kioussis,§ Shu-Yu Ning,∥ Haibin Su,∥ † Toshiaki Itaka,¶ Takami Tohyama,∥ Xinyu Yang,∥ and Jiu-Xing Zhang†, †

†School of Materials Science and Engineering, Hefei University of Technology, Hefei 230009, Anhui, China
‡Department of Physics, California State University Northridge, Northridge, California 91330-8268, USA
§Division of Materials Science, Nanyang Technological University, 50 Nanyang Avenue, 639798 Singapore
¶Institute of Advanced Studies, Nanyang Technological University, 60 Nanyang View, 639673 Singapore
∥Computational Astrophysics Laboratory, RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

(Dated: October 26, 2015)

Using first-principles calculations, we predict a Chern insulating phase in thin films of the ferromagnetic semi-metal GdN. In contrast to previously proposed Chern insulator candidates, which mostly rely on honeycomb lattices, this system affords a great chance to realize the quantum anomalous Hall effect on a square lattice without either a magnetic substrate or transition metal doping, making synthesis easier. The band inversion between 5d-orbitals of Gd and 2p-orbitals of N is verified by first-principles calculation based on density functional theory, and the band gap can be as large as 100 meV within GdN trilayer. With further increase of film thickness, the band gap tends to close and the metallic bulk property becomes obvious.

PACS numbers: 73.43.-f, 71.20.Mq, 73.61.Ey

Topological insulator (TI), as an exotic quantum state has been experimentally realized in several materials with strong spin-orbital coupling.1–3 Chern insulator, as a material which can realize the quantum anomalous Hall effect (QAHE) also have attracted lots of research interest. The QAHE, i.e., quantum Hall effect without external magnetic field, proposed by Haldane on the hexagonal lattice,4 is realized in the chromium doped (Bi,Sb)2Te3 thin-film recently.5–7 However, the chromium-doped (Bi,Sb)2Te3 thin-film exhibits ferromagnetic (FM) spin order only below 15 K, and the transverse conductivity $\sigma_{xy}$ becomes quantized to better than $1/10$ only below 400 mK. In addition, the QAHE also is proposed in the magnetic quantum-well (QW),8–10 honeycomb materials with induced ferromagnetism (FM),11–17 magnetically doped thin-film topological crystalline insulator.18 However, for the magnetic quantum well and honeycomb materials, the band gap usually is very small, and all the proposed materials are difficult to fabricate, more or less. Recently, Chern insulator with square lattice also are proposed in interface of CrO2 and TiO2,19, double-perovskite monolayers20 and the magnetic rocksalt interface21,22. The band gap of EuO/GdN multilayer even can be as larger as 0.1 eV,22 in which the QAHE can persist up to room temperature. The QW structure of Weyl semimetal (WSM), affords another simple platform to realize the QAHE.23 The WSM is a type of topological matter with pairs of separated Weyl points or band crossing. The Weyl points can only appear when the spin-doublet degeneracy of each band is removed by broken time reversal symmetry or spatial inversion symmetry.24 For the realization of QAHE, we can consider the thin-film of WSM with broken time-reversal symmetry resulting from FM spin order. The Weyl semimetal phase in the three dimensional (3D) structure of stacked FM multilayer of TI also was proposed by Burkov et al.25.

In this work, we propose the realization of QAHE in the GdN [001] thin-film, which has a relatively large band gap for working at room temperature. Bulk GdN with rock-salt structure is known as ferromagnetic half-metal with Curie temperature $T_C \sim 58$ K.26–32 It was intensively studied in 1960s and 1990s due to its potential application in spintronics device. By first-principles calculation based on density-functional theory (DFT) and tight-binding (TB) model calculation, we predict that the band crossing in the FM GdN bulk is unavoidable even the spin orbitals coupling (SOC) is included in the first-principles calculation, and the band crossing between $d$-orbitals in GdN bulk is protected by the four-fold rotation symmetry. The $d$-band inversion or crossing has been discovered in several materials, such as non-centrosymmetric superconductor PbTaSe2,33 non-Kondo like topological insulator YbB6,34,35 topological insulator LaX (X=P, As, Sb, Bi), Weyl semimetal TaAs,36 and topological semimetal LaN.37 We also predict FM GdN thin-film as Chern insulator with $d$-band inversion on square lattice. Since the time-reversal symmetry is broken by the FM spin order of Gd 4f-electrons, no magnetic doping or substrate is required. The $d$-band inversion starts in the free standing GdN bilayer, and the the band gap has a maximal value ~100 meV in trilayer. With the increase of thickness of thin-film, the metallic bulk character will become obvious, viz. band gap will approach to zero.

The DFT calculations employed the all electron, full-potential linearized augmented plane wave (FPLAPW) method with generalized gradient approximation plus Hubbard $U$ (GGA+$U$) implemented in WIEN2k code.38 The SOC was included in the self-consistent calculations. We have used the experimental value $a_0=4.97$ Å for the lattice parameter, and the effective Hubbard $U_{eff}$ for the
Gd 4f-orbitals is set to be 9.0 eV, determined by the occupied Gd 4f levels in GdP by optical experiment.\cite{28}

**Bulk GdN** GdN is the unique FM material in the gadolinium monopnictides, and its lattice dependent Curie temperature can be up to 70 K. The electronic structure of FM bulk GdN calculated by GGA+U without SOC is shown in Fig. 1a. With deeply buried 4f-electron resulting from strong electronic correlation and FM Zeeman field, the Gd d_{xy}- and N p-orbitals make the main contribution to the electronic structure near the Fermi level. It reveals that FM bulk GdN is half-metallic, i.e., metallic character within spin-up bands and insulating character within spin-down bands. Of interest, there is a band crossing along the Γ→X direction between Gd d_{xy}-orbitals and N p_{x}(p_{y})-orbitals. This d_{xy}/ p_{x}(p_{y}) band crossing is protected by the M_{zz} (M_{yz}) mirror plane, which maps y(x) to -y(-x). In the Brillouin zone (BZ) (shown in the inset of Fig. 1b), bands on the k_{y}=0 and k_{z}=0 planes can be labeled by their eigenvalues of M_{xx} and M_{yz}, respectively. Obviously, the p_{x}(p_{y})-orbitals and d_{xy}-orbitals have different M_{zz} (M_{yz}) eigenvalues 1, and -i, respectively, and they can’t hybridize on the k_{y}(k_{z})=0 plane. Along the Γ→X direction, i.e., (0,0,0)-(0,0,2π), both p_{x} and p_{y}-orbitals can’t hybridize with the d_{xy}-orbitals. In the TIs, SOC can remove this band crossing and open a gap. With the spin quantization axis along z-axis, p_{x}±iπp_{y}-orbital has quantized orbital momentum h, and it has definite eigenvalue of ±i under four-fold rotation symmetry C_{4z}. However, d_{xy}-orbital has eigenvalue of -1 under C_{4z}.\cite{30} The band crossing between d/p orbitals still can be protected by the four-fold rotation symmetry C_{4z}. Since the high-symmetry line Γ→X is a C_{4z} invariant line, energy bands with different C_{4z} eigenvalue along this line can’t hybridize with each other. So, we conclude that the d/p band crossing in FM bulk GdN is stable against SOC perturbation if the C_{4z} is not broken. We note that the bandwidth of Gd 5d-orbitals is strongly dependent with the lattice constant.\cite{40-44} With reduced lattice parameter, both the bandwidth of Gd 5d and the ferromagnetism will be enhanced in strained GdN.\cite{44}

The band crossing in the FM bulk GdN can be described by a simple tight-binding model (TB) on face-centered cubic lattice with Gd d=d_{xy} and N p=p_{x}±ip_{y} as basis, which can be expressed as,

\[
H = H_{d} + H_{p} + H_{dp},
\]

\[
H_{d} = \epsilon_{d} + t_{d} \sum_{i} d_{i}d_{i+\hat{x}} + t'_{d} \sum_{i} d_{i}d_{i+\hat{y}} + h.c.
\]

\[
H_{p} = t_{p} \sum_{i} p_{i}p_{i+\hat{x}} + t'_{p} \sum_{i} d_{i}d_{i+\hat{y}} + h.c.
\]

\[
H_{dp} = t_{dp} \sum_{i} (d_{i+\hat{x}} - i_{i} + d_{i+\hat{y}} - i_{i}).
\]

With Fourier transformation, the Hamiltonian reads

\[
H = \sum_{k} \left( \begin{array}{c} p_{k} \\ d_{k} \end{array} \right)^{\dagger} \left( \begin{array}{cc} A(k) & V(k) \\ V(k)^{\dagger} & B(k) \end{array} \right) \left( \begin{array}{c} p_{k} \\ d_{k} \end{array} \right)
\]

\[
A(k) = 4t_{d}cos \frac{k_{x}}{2}cos \frac{k_{y}}{2} + 4t'_{d}(cos \frac{k_{y}}{2}cos \frac{k_{z}}{2} + cos \frac{k_{x}}{2}cos \frac{k_{z}}{2})
\]

\[
B(k) = \epsilon_{d} + 4t_{d}cos \frac{k_{x}}{2}cos \frac{k_{y}}{2} + 4t'_{d}(cos \frac{k_{y}}{2}cos \frac{k_{z}}{2} + cos \frac{k_{x}}{2}cos \frac{k_{z}}{2})
\]

\[
V(k) = 2t_{dp}(sin \frac{k_{x}}{2} + i sin \frac{k_{x}}{2})
\]

in momentum space. The A(k) and B(k) terms describe the d- and p-band dispersion without hybridization. The V(k) describes the hybridization on different momentum points. The vanishing d-p hybridization along (0,0,0)-(0,0,π) direction is obvious, and the d-p band crossing at k_{z}=0.69π, estimated by on-site energy \(\epsilon_{d}=4.2\) eV and hopping parameters \(t_{d}=-0.93\) eV, \(t'_{d}=0.23\) eV, \(t_{p}=0.14\) eV, \(t_{p}^{''}=0.07\) eV, \(t_{dp}=0.93\) eV extracted from Wannier calculation with five Gd d-orbitals and three N p-orbitals.\cite{45} With these parameter, the first Chern number is -1 when k_{z} in \((-\pi,-0.69\pi)\) and \((0.69\pi,\pi)\), and zero when k_{z} in \((-0.69\pi,0.69\pi)\). The calculated band structures and surface states of GdN by TB model are provided in the supplementary file.\cite{46}

**Thin-film GdN** Since the Gd d_{xy} and N p band crossing is protected by the four-fold rotation symmetry C_{4z}, it’s possible to remove this band crossing by size confinement, i.e., eliminate the z-direction. The GdN [001]

![Image](image.png)
thin-film, which can be regarded as a stack of GdN A and B monolayer along the z-direction, becomes a prospective Chern insulator candidate with square lattice, instead of the extensively studied honeycomb lattice. With lattice vectors $\vec{a} = \frac{a_0}{\sqrt{2}}(1,0)$, $\vec{b} = \frac{a_0}{\sqrt{2}}(0,1)$, B monolayer has a offset (0.5,0.5) relative to the A monolayer. With vanishing inter-layer hopping parameter $t'_d = t'_p = 0$ in the TB model, the $d_{xy}$ band always has higher energy than the $p$-orbitals dominating bands over the whole BZ, i.e., the monolayer of GdN should be insulating, which is consistent with our first-principles calculated result of free standing GdN monolayer. The calculated electronic structures of free standing AB bilayer (BL), ABA trilayer (TL) and 4 monolayers with thickness $\lambda = a_0$, 1.5$a_0$, and 2$a_0$ by first-principles calculation with GGA+$U_{eff}$+SOC are shown in Fig. 2. The spin-up Gd $d_{z^2-\gamma^2}$-orbitals in red and N $p(=p_x \pm ip_y)$-orbitals in blue are hybridized near the Fermi level, and all the spin-down bands are in black. Our calculated results show that the $d/p$ band inversion starts in the BL, though the band gap resulting from $d$-$p$ hybridization is minute. However, in the TL, the band gap can increase up to 100 meV, which almost is the same as the band gap in GdN/EuO interface. Because of the enhanced bandwidth resulting from interlayer $d$-$d$ and $p$-$p$ hybridization, the $d$-orbitals and $p$-orbitals on the B layer are inverted near the center of the BZ and hybridized and this $d/p$ band inversion contributes -1 to the Chern number. In both GdN bilayer and trilayer, the A monolayer behaves like a normal 2D insulator because there is no $d$-band from A layer involving with the band inversion as shown in Fig. 2b. This GdN trilayer can be regarded as Chern insulator sandwiched by ordinary insulator layer. The intra-layer $d/p$ hybridization gap is $\sim 0.3$ eV. However, the global band gap is reduced to 100 meV because of the weaker inter-layer $d/p$ hybridization. With further increase of thickness, more $d/p$ band inversions trend to take place. However the band gap is seriously reduced because of the vanishing long distant inter-layer $d/p$ hybridization, as shown in Fig. 2c. Even though the intra-layer $d$-$p$ hybridization can be very strong here, the $p$-bands from the normal insulator layers always are obstructions to obtain large band gap. We also notice that the calculated bang gap also decreases with the increase of GdN monolayer in EuO/GdN multilayer.

The electronic structure of one dimensional ribbon of TL is calculated by the TB model with Gd $d_{z^2-\gamma^2}$-orbitals and N $p_x + ip_y$-orbitals as basis with on-site energy $\epsilon_d = 4.2$ eV and $t_{dd} = -0.73$ eV, $t'_{dd} = 0.23$ eV, $t'_{pp} = 0.07$ eV, $t_{dp} = 0.93$ eV.$t'_{dp} = 0.05$ eV. A pair of metallic surface state comes from the left and right termination of ribbon.

For the epitaxial fabrication of [001] GdN thin-film, usually a substrate is required. Here we also calculate the electronic structure of GdN TL with six monolayers of YN. The lattice parameter of YN in rock-salt structure is 4.88 Å, which is very close to the GdN lattice parameter 4.98 Å. The electronic structure of this GdN/YN multi-

![FIG. 2: (Color online) Band structures of GdN (AB) bilayer (a), (ABA) trilayer (b), and 2 bilayers (c), calculated by GGA+$U_{eff}$+SOC. The weight of $d_{z^2-\gamma^2}$ and $p_x+p_y$ orbitals is represented by red and blue colors. All the spin-down bands are in black.](image1)

![FIG. 3: Calculated tight-binding electronic structure of FM GdN in form of 1D ribbon with $\epsilon_d = 4.2$ eV, $t_{pp} = 0.14$ eV, $t_{dd} = -0.73$ eV, $t'_{dd} = 0.23$ eV, $t'_{pp} = 0.07$ eV, $t_{dp} = 0.93$ eV, $t'_{dp} = 0.05$ eV. A pair of metallic surface state comes from the left and right termination of ribbon.](image2)
layers calculated by GGA + $U_{eff}$ + SOC is shown in Fig. 4 with Gd $d_{x^2-y^2}$ orbitals in red, and the $d/p$ band inversion is reproduced. However, the calculated band gap is about 20 meV, indicating a better substrate should be explored in future work.

In summary, we predict the Weyl semimetal phase and Chern insulator phase in FM bulk and thin-film GdN, respectively. In the bulk GdN, the $d$-$p$ band crossing is protected by the four-fold rotation symmetry C$_4$, and it is robust against SOC. Chern insulating state on square lattice is predicted on the [001] thin-film GdN, and its multilayer structure affords a concrete realization of 3D Weyl semimetal phase. The maximal band gap is $\sim$100 meV, which presents in the free standing GdN TL with a single $d/p$ band inversion. With the increasing thickness of thin-film, the band gap trends to close and metallic bulk state will become obvious.

This work is supported by National Natural Science Fund of China under No. 51371010 and the Fundamental Research Funds for the Central Universities. The research at CSUN was supported by NSF-PREM Grant No. DMR-1205734. Authors also are grateful to J. P. Hu for useful discussion.

FIG. 4: Calculated electronic structure of GdN TL on six YN monolayers by GGA + $U_{eff}$ + SOC. The Gd $d_{x^2-y^2}$ orbitals are in red.


1 M. Z. Hasan and C. L. Kane, Rev. Mod. Phys. 82, 3045 (2010).

39 $d_{xy} \pm id_{x^2-y^2}$ and $d_{xy}$ have the same $C_4z$ eigenvalue.

There’s no difference to choose $d_{xy}$ or $d_{xy} \pm id_{x^2-y^2}$ as basis vector.

39 $d_{xy} \pm id_{x^2-y^2}$ and $d_{xy}$ have the same $C_4z$ eigenvalue.

There’s no difference to choose $d_{xy}$ or $d_{xy} \pm id_{x^2-y^2}$ as basis vector.

46 See Supplemental Material at [URL will be inserted by publisher] for the calculated band structures and surface states of GdN.