Quantum spin Hall effect in IV-VI topological crystalline insulators

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Abstract

We envision that the quantum spin Hall effect should be observed in (111)-oriented thin films of SnSe and SnTe topological crystalline insulators. Using a tight-binding approach supported by first-principles calculations of the band structures, we demonstrate that in these films the energy gaps in the two-dimensional band spectrum depend in an oscillatory fashion on the layer thickness. These results as well as the calculated topological invariant indexes and edge state spin polarizations show that for films ∼20–40 monolayers thick a two-dimensional topological insulator phase appears. In this range of thicknesses in both SnSe and SnTe, (111)-oriented films edge states with Dirac cones with opposite spin polarization in their two branches are obtained. While in the SnTe layers a single Dirac cone appears at the projection of the $\Gamma$ point of the two-dimensional Brillouin zone, in the SnSe (111)-oriented layers three Dirac cones at $M$ points projections are predicted.

1. Introduction

Topological insulators (TIs) and the quantum spin Hall (QSH) effect attract significant interest for both fundamental and practical reasons. In the three-dimensional (3D) and two-dimensional (2D) TIs the bulk insulating states are accompanied by metallic helical Dirac-like massless electronic states on the surface (edges) of the crystal. Due to the time-reversal symmetry, these surface states are topologically protected against scattering at 180°. In 2D TIs this means that the metallic edge states provide dissipationless, spin-polarized conduction channels. Such property makes these structures extremely interesting for low-power-consumption electronics and spintronics. Thus, the quest for systems with topologically non-trivial edge states, which give rise to the QSH effect, has become recently one of the most important topics in condensed matter physics.

The QSH effect is the net result of two opposite polarized spin currents traveling in opposite directions along the edges of a 2D TI. It has been predicted by Kane and Mele in graphene [1]. In this material, however, it could occur only at unrealistically low temperatures, since the intrinsic spin-orbit coupling, which should open a band gap at the Dirac points, is very weak. Subsequently, it has been proposed and confirmed experimentally that the QSH state might arise in HgTe/HgCdTe quantum wells [2] and also in InAs/GaSb heterostructures [3]. In these experiments the transition from insulating to conducting behavior has been observed. In both cases the conductivity was close to the $2e^2/h$ value expected for the two parallel quantum Hall channels. The spin polarized nature of the edge states has been shown much later and only in the HgTe-based structure [4]. All these observations were made at very low temperatures, below 10 K, due to the small energy gap in these systems.

In the search for new QSH structures many theoretical predictions have been made, in which various, sometimes exotic, chemical classes of 2D materials were considered. These include a slightly buckled honeycomb lattice of Si atoms (silicene) [5], Bi bilayers on different substrates [6], Ge(Bi$_{0.5}$Sb$_{1-x}$)$_2$Te$_4$ with various Bi concentrations [7] and functionalized ultrathin tin films (stanene) [8]. Recently, it has also been shown that 2D transition metal (Mo or W) dichalcogenides should form a new class of large-gap QSH insulators [9]. Finally, a 2D TI with an energy gap as large as 0.8 eV has been predicted for an overlayer of Bi grown on a semiconductor Si(111) surface functionalized with a one-third monolayer of halogen atoms [10].
these theoretical predictions, no stand-alone thin film or a thin film supported on a suitable substrate have been up to now experimentally demonstrated to harbor a QSH state.

2. Results and discussion

In this article we describe a study of (111)-oriented thin films of well-known IV-VI semiconductors SnSe and SnTe, which can be quite easily grown on a BaF₂ substrate by, e.g., molecular beam epitaxy. Such layers have a rock–salt crystal structure, despite the fact that bulk SnSe has an orthorhombic structure [11]. Recently, it has been shown that these compounds are so-called topological crystalline insulators (TCIs). TCIs are non-trivial insulators supporting surface Dirac fermions protected not by time reversal but by crystal symmetry [12]. The IV-VI semiconductors, i.e., SnSe and SnTe, as well as PbTe and PbSe and the substitutional solid solutions of Pb- and Sn-based chalcogenides, were identified earlier as trivial insulators. This is because in these compounds the band inversion happens simultaneously at an even number (four) of L points in the Brillouin zone (BZ). The angle-resolved photoemission spectroscopy (ARPES) confirmed, however, that these materials belong to the TCI class. It has been shown that metallic surface states exist on the (001) surfaces of SnTe [13] as well as Pb₁₋ₓSnₓSe [14] and Pb₁₋ₓSnₓTe [15]. These gapless (001) surface states are supported by mirror symmetry. It should be emphasized that due to the spin-orbit interactions the metallic surface states observed in real SnTe- or SnSe-based compounds have almost linear, Dirac-like dispersions. In (001)-oriented thin films of TCIs hybridization leads to a new 2D TCI phase, which supports two pairs of spin-filtered edge states [16, 17]. These edge states are protected solely by mirror symmetry—the topological phase is indexed by the mirror Chern number |N_M| = 2. In contrast, we note that in principle the 2D TCI phase is not expected in (111)-oriented films. Our calculations show, however, that in ultrathin films (SnTe and SnSe) grown not along the (001) but along the (111) crystallographic axis, a QSH state can be obtained. This is because the four L point projections onto the 2D BZ of a (111)-oriented thin film are not equivalent. While the energy structures at the three M̅ projections are the same by symmetry, the energy structure at the fourth projection at the Γ̅ point is different. This results in a possibility that the band inversion takes place in an odd (either one or three) number of points.

A IV–VI crystal in a rock–salt structure has [111] planes alternately composed of either cations or anions. Thus, (111)-oriented slabs consisting of an even number of monolayers have one surface cation- and the other anion-terminated. In contrast, in slabs with an odd number of layers both surfaces are the same and the inversion symmetry is preserved. These two cases have to be distinguished.

2.1. Odd number of monolayers

For a (111)-oriented SnTe or SnSe thick film four single, topologically protected Dirac cones in the four projections of the L̅ points onto the 2D BZ (one in the Γ̅ point and three in the M̅̅̅̅̅̅ points) are obtained in the calculations. This is similar to what we have obtained before for a (111)-oriented bulk PbSnTe crystal [18]. For the anion-terminated surfaces, the bands are brought to contact forming anion Dirac cones, while in the other case the bands meet to form cation Dirac cones. As shown already in [18, 19], all L points belong to the three {110} mirror planes of the (111) surface and all Dirac points should be thus topologically protected. These predictions have been also confirmed experimentally for (Pb,Sn)Se [20] as well as for SnTe [21]. While at the Γ̅ point the Dirac cone is isotropic, the band structure around M̅ is strongly anisotropic, i.e., depends differently on the k̅ values along the M̅−Γ̅̅ and M̅−K̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅̅...
of the band gap result from the $k^2$ corrections to the Dirac-like Hamiltonian of massive electrons in the direction perpendicular to the surface and have been already predicted for thin films of 3D TIs, like Bi$_2$Se$_3$ or Bi$_2$Te$_3$ [22, 23]. The differences in the results obtained for SnTe and SnSe films stem from the fact that in these materials $k^2$ terms are highly anisotropic. Namely, in SnTe the $k^2$ term moves the energy gap from $L$ towards the $\Gamma$ point, whereas in SnSe, in directions perpendicular to the $L - \Gamma$ line (see figure S3 in the supplementary material). Indeed, as shown in the face centered cubic (fcc) BZ schematic (figure S4) for $L_1$ the direction perpendicular to the (111) surface follows the $L_1 - \Gamma$ line, while for the other $L$ points it is inclined at a small angle to the ($W$-$L$-$K$) plane.

Analogous to the results obtained in [17] for (001)-oriented Pb$_{1-x}$Sn$_x$Te layers, the described changes of the energy gaps are associated with a topological phase transition. Our calculations show, however, that in the case of (111)-oriented SnTe and SnSe layers the topological phase changes from trivial insulator not to TCI, but to the 2D TI. The effect is similar to the transition to 2D TI predicted for Bi$_2$Se$_3$ or Bi$_2$Te$_3$ thin films [23]. This result is obtained by calculations of the $Z_2$ invariant for the four time-reversal momenta points in the BZ (one $\Gamma$ and three $\overline{M}$) determines the $Z_2$ invariants [24].

In figures 2(a) and (b) we present the calculated [110] edge spectral functions of the (111)-SnTe cation-terminated film consisting of 19 monolayers (i.e., with inverted band gap as shown in figure 1(a)). The results for a slab consisting of 13 monolayers, where a trivial insulator phase is expected, is presented in figure 2(c). The 2D BZ zone of the (111)-oriented film and 1D BZ of the [110] edge are presented in figure 2(d). As shown in the latter, the $\Gamma$ and $\overline{M}_1$ points project onto the edge at $k_x = 0$, while $\overline{M}_2$ and $\overline{M}_3$ at $k_x = \pm \sqrt{2} \pi/\alpha_0$. In agreement with the results of the topological invariant calculations, shown earlier, Dirac crossing of the edge states appears in the band gap at the $\Gamma$ projection for the 19-layer, but not for the 13-layer-thick slab (compare figures 2(a) and (c)). The projection of $\overline{M}_2$ and $\overline{M}_3$ (for $k_x = \sqrt{2} \pi/\alpha_0$) the Dirac cones are not obtained for any SnTe layer thickness. In contrast, in SnSe, as shown in figures 3(a) and (b) for the anion-terminated 21-monolayer thick
slab, two Dirac crossings of the edge states appear inside the band gap at the $k_z = \sqrt{2} \pi/a_0$ point. As the third $M_1$ point projects again at $k_x = 0$ where thus another edge state with a Dirac node appears. Due to the strong band overlapping, the latter is dispersed within the valence band.

Finally, we have calculated the $G^\uparrow$ and $G^\downarrow$ contributions of the spin-up and spin-down (111) projections in the edge spectral functions. The sign of the $G^\uparrow - G^\downarrow$ difference corresponds to spin polarization of the edge states. As expected for the 2D TI in the QSH state, the obtained edge states are spin polarized and the spin polarizations in the two branches of the Dirac cones are always opposite. These spin polarizations in all studied cases are about 80%, i.e., the contribution of the dominant spin configuration surpasses ca 8–10 times the opposite spin orientation.

2.2. Even number of monolayers

Unlike the case of an odd number of layers, for (111)-oriented slabs consisting of an even number of monolayers there is no inversion symmetry in the system and the energy gaps are moved from the time-reversal invariant points (see figures S1(b) and S2(b) in supplementary section I). The gap inversion occurs now in two points related by the time reversal symmetry on two sides of each time reversal invariant $k$ point. It has been shown for inversion-asymmetric systems in general [25] that the gap closing can be accompanied by a transition between the trivial insulator and the QSH phase when the gap inversion occurs in an odd number of such pairs. In SnSe films this takes place in three pairs in the vicinity of the three nonequivalent $M$ points while in SnTe around the $T$ point. Our calculations show that indeed for a given range of thicknesses the topological phase transition to the TI phase can be expected in these (111)-oriented films.

![Figure 2](image-url) Edge states in cation-terminated (111)-SnTe film. The calculated spectral functions for a slab of 19 monolayers at the projection of (a) $\Gamma$ and (b) $T_M$ and $M_1$ points onto the [110] edge. (c) The spectral function at the projection of $\Gamma$ for 13-monolayer-thick film. (d) The 2D BZ of (111)-oriented film (green hexagon) and 1D BZ of its [110] edge (red line). Local extrema of the 2D bands projected to the edge are here and in the following figures denoted by black dotted lines.
If there is no inversion symmetry in the system, the $Z_2$ topological invariants can not be determined by the method utilizing the parity of the eigenfunctions, which we used for films with an odd number of monolayers. To calculate $Z_2$ for such layers we have adopted the method proposed by Fukai and Hatsugai [26]. In this method the improvements to computing Chern numbers in a lattice BZ enable the tight-binding calculations of $Z_2$ based on the formula derived by Fu and Kane in [27]. The dependence of the band gap of SnTe (111)-oriented films consisting of an even number of monolayers on the layer thickness is shown in figure 4(a). The thicknesses for which the value $\nu = 1$ has been obtained are marked by the green shadow. As one can see in figure 4(a) the QSH phase can be expected in SnTe films consisting of 14–28 monolayers (\sim 2.5–5.1 nm thick). For SnSe the QSH phase is obtained for 18–38 monolayers, i.e., for film thicknesses in the range 3.1–6.6 nm. In the case of SnTe films with an even number of layers the whole inverted energy gap for states in the vicinity of $\Gamma$ is situated inside the large gap between conduction and valence bands close to the $M$ point, as one can clearly see in figure 4(b). The largest value of the inverted gap, about 75 meV, is obtained for the 18–20-monolayer-thick SnTe slab.

The calculated spectral functions along the [110] edge for the (111)-oriented, 18-monolayers-thick SnTe film show a clear Dirac node in the center of the band gap at the $k_x = 0$ point. This is shown in figure 5(a). The obtained results are in full agreement with the formalism developed in [25]. We note that a similar result has been obtained for graphene [1]—in this case one observes a single edge state with a Dirac node at $k_x = 0$ (time-reversal symmetry point), despite the energy gaps appearing in two points $K$ (in which the time reversal is not preserved).

**Figure 3.** Edge states in 21-monolayers-thick anion-terminated (111)-SnSe film. The edge spectral functions in the vicinity of (a) $k_x = 0$ (a) and (b) $k_x = \sqrt{2} \pi/a_0$ points of the 1D BZ. The corresponding spin densities are shown on the right. Red and green lines represent the spin-down and spin-up polarization, respectively.
Figure 4. Energy gaps in SnTe film with an even number of monolayers. (a) The dependence of the 2D state band gaps on the slab thickness. For thicknesses within the green shadowed area $v = 1$ was obtained. (b) The valence and conduction bands extrema in the vicinity of $\Gamma$ (black) and $M$ (red) versus the number of monolayers.

Figure 5. Edge states in 18-monolayer-thick SnTe film. (a) The edge spectral functions at the vicinity of $k_x = 0$ point of the 1D BZ. (b) The corresponding spin density. The spin-down polarization is denoted by red, the spin-up by green color.
Figure 6 presents the edge spectral functions at the projections of $\mathbf{K}$ points at (a) $k_x = 0$ and (b) $k_x = \sqrt{2} \pi/a_0$ point in (b). The spin polarizations of the edge states presented in (a) and (b) are shown in (c) and (d), respectively. Again, green color represents the spin-up and red represents the spin-down contribution to the edge spectral function.

Figure 6 presents the edge spectral functions at the projections of $\mathbf{K}$ points at (a) $k_x = 0$ and (b) $k_x = \sqrt{2} \pi/a_0$ point, calculated for the 20-monolayer-thick SnSe slab. As shown in the Supplementary Material, in this case the band gap at the $k_x = 0$ is very small. Still, a pair of edge states can be seen near the top of the valence band. The spin polarization of this edge state is shown in figure 6(c). At the $k_x = \sqrt{2} \pi/a_0$ point, however, the edge states that form the two Dirac cones do not cross over the band gap to connect the valence and conduction bands. Instead, the upper and lower edge states repel each other and two anticrossings appear (see figure 6(b)).

We recall that this is in contrast to the result obtained for films with an odd number of monolayers, where two Dirac cones intersect. There is no crystal symmetry which can protect such additional Dirac points. They cannot be explained by the inversion symmetry, which distinguishes the two cases, because an edge in the semi-infinite sample breaks this symmetry. Surprisingly enough, we observe that the same difference between the odd and even cases can be obtained within a simple effective mass model. The Hamiltonian $\hat{H}_0 = v k_x \hat{s}_x - v k_y \hat{s}_y$ proposed in [28] is used to describe the surface states. For two surfaces of a thick film, the Hamiltonian has the form:

$$\hat{H} = (\hat{H}_0 + \Delta) \hat{\tau}_z$$

where $\tau_z = \pm 1$ describes the upper and lower surface, respectively, and $s_z = \pm 1$ denotes a Kramers doublet. $2\Delta$ is the energy difference between the Dirac point positions for anionic and cationic surfaces and equals zero when the upper and lower surfaces are the same. For films with an odd number of monolayers this effective Hamiltonian is invariant not only against the crystalline symmetries of the system but also against reflection from the mirror plane situated in the middle of the film, $\hat{M}_z = -i s_z \hat{\tau}_z$. When the thickness of the film decreases
the states of the two surfaces start to interact. This interaction, which is described by the \( M_{\tau_x} \) operator does not break the mirror symmetry. The conclusion is valid for any \( L \)-point projection. The \( [110] \) edge mixes \( \Gamma \) with \( M_1 \) and \( M_2 \) with \( M_3 \), but also does not destroy the mirror plane symmetry. In case of films with an odd number of monolayers this symmetry protects the Dirac cone intersections. Moreover, it provides additional protection of Dirac points in the time-reversal invariant momenta. In contrast, such symmetry does not exist in films consisting of an even number of layers, with two different surfaces, i.e., with a nonzero \( \Delta_{\tau_z} \) term in the Hamiltonian \( H \). Thus, for such SnSe films instead of additional Dirac points, anticrossings appear (see figure 6).

3. Conclusions

The calculations presented in this paper show that in the free-standing \((111)\)-oriented SnSe and SnTe thin films there exists a range of thicknesses for which the 2D TI phase appears. The QSH phase is obtained for all studied films, both with odd and even numbers of monolayers. Moreover, in SnSe films with an odd number of monolayers, for thicknesses for which the 2D TI phase is predicted, additional Dirac points appear out of the time-reversal symmetry points. Within an effective mass description, in this case the QSH phase coexists with a phase that can be called a ‘2D effective TCI’ and the topology is protected by both time-reversal and mirror plane symmetries. For SnTe films with an odd number of monolayers this is also the case but only for one Dirac point at \( k = 0 \). The model allows us also to calculate the appropriate mirror Chern numbers—ones obtains \( |N_M| = 1 \) in SnTe films while \( |N_M| = 3 \) in SnSe.

However, for all but the \((111)\)-oriented SnTe films with an even number of monolayers an overlapping of bands in \( \hat{\mathcal{T}} \) and \( \hat{\mathcal{M}} \) diminishes the final band gap. Hence, the edge states appear either against the background of the bands or within a very small energy gap (see supplementary section II). The \((111)\)-oriented SnTe films with an even, close to 20, number of monolayers is thus the best candidate for observing the QSH effect in the IV-VI TCI. The QSH phase produced in this material system is highly robust, being even oblivious to the lack of inversion symmetry. The latter indicates that a suitable substrate or an overlayer will not destroy this 2D TI phase. Quite the contrary, using carefully selected substrate or overlay material (e.g., \( Pb_{1-x}Eu_{x}Se_{1-y},Te_{y} \) quaternary alloy) would allow us to tune the height of the energy barriers and biaxial strain in the structure [29]. As shown already in [30] for SnTe (001) surface and for SnTe nanomembrane in [31], the uniaxial or biaxial strain can be used for manipulating the energy gaps in the surface (or interface) states. The strain can be also used for solving the mentioned problem of band overlapping, which diminishes the resulting energy gap (as it was shown in [32] for solving a similar obstacle in PbTe/(Pb,Sn)Te heterostructures). Thus, these results may pave the road for the experimental realization of QSH in SnTe and SnSe layers and their quantum well structures.

Note added: after the arXiv preprint of this paper appeared, similar results for SnTe layers in the presence of an electric field have been published [33].

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