Spin Chirality Tuning and Topological Semimetals in Strained HgTe$_x$S$_{1-x}$

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By means of detailed electronic structure calculations, we show that strained HgTe$_x$S$_{1-x}$ alloys show a surprisingly rich topological phase diagram. In the strong topological insulator phase, the spin chirality of the topological nontrivial surface states can be reversed by adjusting the alloy concentration $x$ and the strain. On top of this, we predict two semimetallic topological phases, namely, a Dirac semimetal and a Weyl semimetal. The topological phases are characterized by their $Z_2$ invariants and their mirror Chern numbers.

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Introduction.—Topological insulators (TIs) are bulk insulators that possess a topologically nontrivial band structure. As a consequence, these materials host spin-polarized and topologically protected surface states that cross the fundamental band gap [1,2]. Experimental and theoretical investigations have focused on $Z_2$ TIs and on topological crystalline insulators (TCIs). The former exist if time-reversal symmetry is unbroken; they are characterized by the $Z_2$ topological invariant $\nu$ and show an odd number of band inversions at time-reversal invariant momenta in the bulk Brillouin zone [3–5]. Probably the most prominent three-dimensional TIs in this class are the Bi chalcogenides, e.g., Bi$_2$Se$_3$ and Bi$_2$Te$_3$ [6]. Topological crystalline insulators, on the other hand, rely on unbroken crystal symmetries [7], typically a mirror symmetry. A prominent topological crystalline insulator is SnTe [8]. The relevant topological invariant is the mirror Chern number. Topologically nontrivial surface states show up on any surface perpendicular to the mirror plane. Their Dirac points are then located on the line that is the intersection of the mirror plane with the surface Brillouin zone (BZ). It turned out that some compounds fall into both classes; for example, Bi$_2$Te$_3$ is both a TI and a TCI [9].

A mirror Chern number provides information on both the number and the spin chirality of the topologically nontrivial surface states. More precisely, its modulus gives the number of topologically nontrivial surface states, whereas its sign tells whether their spin chirality is clockwise or anticlockwise in the upper part of the Dirac cone [5]. Topological insulators investigated so far have a clockwise spin chirality, which is explained by the orbital composition of the surface states: the Dirac cones are composed mostly of orbitals aligned normal to the surface (e.g., $p_z$ orbitals) [10]. In contrast, surface-parallel orbitals (e.g., $p_x$ and $p_y$) favor anticlockwise chirality. Furthermore, one can attribute a positive (negative) spin-orbit coupling constant with clockwise (anticlockwise) spin chirality to $p$ orbitals. These considerations immediately suggest studying the properties of TIs with anticlockwise spin chirality [11]. On top of this, it is desirable to tune the spin chirality, which is important for applications.

Among the first systems investigated in this context are HgTe quantum wells sandwiched between CdTe layers [12,13]. Strained HgTe films are three-dimensional TIs with clockwise spin chirality [14–17]. The similar compound HgS is a TI as well [18], even if unstrained, but its spin chirality is opposite to that of HgTe [20–22]; such an effective negative spin-orbit coupling is acquired by hybridization of the $p$ orbitals with energetically close $d$ orbitals [23].

In this Letter, we report on strained alloys with the zinc blende structure. We show theoretically that the spin chirality of HgTe$_x$S$_{1-x}$ compounds can be tuned, that is reversed, by application of moderate strain in the [001] direction, and by changing the composition $x$. Moreover, these materials exhibit a surprisingly rich topological phase diagram that not only shows TCIs but also topological semimetals: a Weyl semimetal [24] and a Dirac semimetal phase [25]. Thus, the topological properties and, consequently, the spin-dependent transport properties of these systems can be adjusted by two external parameters: strain and composition. These findings are important for both device applications and fundamental condensed-matter physics.

Theoretical.—In an initial step, we performed first-principles electronic structure calculations [26] based on the multiple-scattering theoretical formulation of density functional theory (DFT) [34]. We applied the generalized gradient approximation as introduced by Perdew et al. (Ref. [35]). Our homemade Korringa-Kohn-Rostoker computer program package [36–38] accounts for relativistic effects, in particular the essential spin-orbit interaction, by solving the Dirac equation. The lattice constants for HgTe ($6.461$ Å) and HgS ($5.850$ Å) are chosen to match the experimental lattice parameters [39,40]. The DFT band structure of HgTe fits very well to the angle-resolved...
photoelectron spectroscopy measurements \cite{14,41,42} near the $\Gamma$ point of the BZ. Since HgS is very similar to HgTe, we believe that we also get the correct band structure for this case.

Subsequently, tight-binding parametrizations in the Slater-Koster framework \cite{43} were obtained using the DFT results as input. The parameters have been optimized by Monte Carlo simulations \cite{44}. These tight-binding Hamiltonians are then used for all further calculations.

The HgTe$_{1-x}$S$_x$ alloys were simulated by the virtual crystal approximation and the coherent potential approximation (CPA) \cite{45,46}, both giving similar results. A strain $\alpha$ in the [001] direction accompanied by an opposite strain $\beta$ in the in-plane directions was applied; the volume of the unit cell is not conserved. The elastic constants for HgTe and HgS taken from Ref. \cite{47} were linearly interpolated with the concentration $x$.

Both HgTe and HgS crystallize in the zinc blende structure, which lacks inversion symmetry. The (001) surface is terminated either by a Hg or by a Te$_x$S$_{1-x}$ layer. The surface electronic structure has been calculated for semi-infinite systems using Green function renormalization \cite{48}. The dispersion, spin texture, and orbital composition of the surface states are obtained from the layer-resolved spectral density.

For building up a topological phase diagram, we varied the concentration $x$ from 0 to 1 in steps of 0.1 and the strain $\beta$ from 0.97 to 1.03 in steps of 0.01. For all nonmetallic $\beta$ systems we calculated the Chern numbers $c_m$ for the two mirror planes perpendicular to the (001) surface, using the virtual crystal approximation. The phase diagram shows five topological phases, which are discussed in the following (Fig. 1).

**Spin chirality tuning.**—Unstrained HgTe is a semimetal: the BZ center $\Gamma$ the bulk bands at the Fermi level are fourfold degenerate and are inverted; that is, the $\Gamma_6$ band has a lower energy than the $\Gamma_8$ band \cite{15,16}. This degeneracy is lifted by positive in-plane strain ($\beta > 1$), making HgTe a strong TI with $Z_2$ invariant $\nu = (1; 0, 0, 0)$ and a TCI with mirror Chern number $c_m = -1$. Hence, positively strained HgTe is a TI with clockwise spin chirality. The computed surface electronic structure (Fig. 2) fits well to that of Wu et al. (Ref. \cite{17}). Thus, our approach reproduces the topological aspects reported earlier.

HgS is a strong TI with counterclockwise spin chirality \cite{20,21,49}, which is confirmed by our calculations: $\nu = (1; 0, 0, 0)$ and $c_m = +1$. The spectral density of the $S$-terminated (001) surface hosts a topological nontrivial surface state at $\Gamma$ whose spin chirality is opposite to that of strained HgTe (Fig. 2). Application of both positive and negative in-plane strain does not change the topological properties (green area in Fig. 1).

With two similar systems but with opposite spin chirality at hand, one is able to tune—more precisely, reverse—the spin texture of the topologically nontrivial surface state by alloying. The topological phase transition $c_m = -1 \leftrightarrow +1$ is accompanied by a closing of the fundamental band gap and by a vanishing effective spin-orbit coupling. For the unstrained alloy HgTe$_{1-x}$S$_x$ this transition takes place at the critical concentration $x_c \approx 0.15$, for which the $p$ bands are sixfold degenerate at $\Gamma$, indicating a semimetallic system. Calculations performed within the coherent potential approximation instead of the virtual crystal approximation yield an $x_c$ of about 0.20. These numbers set the approximate range for experimental realization of the spin chirality.

**FIG. 1** (color online). Topological phase diagram of strained HgTe$_{1-x}$S$_x$ alloys. Parameters are the concentration $x$ and the strain $\beta$. The phases are indexed by colors: strong TI (STI) with mirror Chern number $c_m = -1$ (red), strong TI with $c_m = +1$ (green), Dirac semimetal (yellow), Weyl semimetal (WSM) (gray), and normal semimetal (blue).

**FIG. 2** (color online). Surface electronic structures of positively strained HgTe (left, $\beta = 1.03$) and unstrained HgS (right), with Te- or S-terminated (001) surfaces, respectively. The spin-resolved spectral density of the topmost layer is shown along a part of the $\bar{Y}$–$\Gamma$–$\bar{Y}$ line of the surface Brillouin zone. The so-called Rashba component of the spin polarization (in plane, perpendicular to the wave vector) is given by the color scale with red positive and blue negative, in states per eV.
reversal symmetry [here \( k \) wave vectors are connected with each other by time-reversal symmetry, forming a pair of Dirac points (Fig. 4)]. The associated system is close to the phase transition. One fifth of the \( x \) spin-orbit coupling almost vanishes at \( \Gamma \), and the \( k_z \) and conduction bands touch at two points on the \( \Gamma \)-\( L \) lines. A further increase of \( x \) shifts the Weyl points within the \( k_z = 0 \) plane towards the \( \Gamma-X \) lines. The four pairs touch immediately at the phase transition; instead, each point at which the valence and the conduction band touch splits into a pair of Weyl points that carry opposite topological charges (Chern numbers \( \pm 1 \)). The associated topologically nontrivial surface states connect the projections of the Weyl points in the bulk onto the surface BZ, leading to so-called Fermi arcs with unique transport properties. These fascinating features are investigated with great effort [50–53].

Upon the application of strain to HgTe\(_{1-x}\), we found Weyl semimetal phases in a region of concentrations with small effective SOC. Starting at \( \beta = 1.03 \) and \( x = 0.0 \) (HgS) in the topological phase diagram (gray area in Fig. 1), a phase transition takes place at \( x_c \approx 0.08 \). There, the valence and conduction bands touch at four points on the \( \Gamma-K \) lines, which are cuts of the \( k_z = 0 \) plane with the \( \{110\} \) or the \( \{1\bar{1}0\} \) mirror plane in the BZ, respectively. Hence, there is a fourfold degeneracy at these points. Upon increasing \( x \), these points split into pairs of twofold degenerate touching points; the bands there disperse linearly, forming Dirac cones. The topological charges associated with these Weyl points are calculated to be \( \pm 1 \).

These bulk properties are nicely supported by the electronic structure of the (001) surface (Fig. 5). For Fermi energy cuts, the spectral densities show Fermi arcs that connect projections of the Weyl points onto the surface BZ. A further increase of \( x \) shifts the Weyl points within the \( k_z = 0 \) plane towards the \( \Gamma-X \) lines. The four pairs touch (but do not recombine because the touching Weyl points possess the same topological charge), exactly for vanishing SOC \( (x_c \approx 0.15) \). For positive SOC \( (x > 0.15) \), the Weyl

FIG. 4. Bulk band structure of unstrained HgTe\(_{1-x}\) for \( x = 0.10 \) (left), \( x = 0.15 \) (center), and \( x = 0.20 \) (right). The spin-orbit coupling almost vanishes at \( x = 0.15 \), and the system is close to the phase transition. One fifth of the \( X-\Gamma \) and the \( \Gamma-L \) lines is shown.

FIG. 3. Bulk band structures of unstrained HgTe\(_{1-x}\), we found Weyl semimetal phases in a region of concentrations with small effective SOC. Starting at \( \beta = 1.03 \) and \( x = 0.0 \) (HgS) in the topological phase diagram (gray area in Fig. 1), a phase transition takes place at \( x_c \approx 0.08 \). There, the valence and conduction bands touch at four points on the \( \Gamma-K \) lines, which are cuts of the \( k_z = 0 \) plane with the \( \{110\} \) or the \( \{1\bar{1}0\} \) mirror plane in the BZ, respectively. Hence, there is a fourfold degeneracy at these points. Upon increasing \( x \), these points split into pairs of twofold degenerate touching points; the bands there disperse linearly, forming Dirac cones. The topological charges associated with these Weyl points are calculated to be \( \pm 1 \).

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FIG. 5 (color online). Electronic structure of the (001) surface of positively strained HgTe\(_{0.11}\)S\(_{0.89}\) with \( \beta = 1.03 \). The spectral density of the topmost layer is given by the color scale (in states per eV). Left: TeS termination. Right: Hg termination. Red dots mark the position of the Weyl points on the surface BZ; their topological charges \( \pm 1 \) are indicated. The arrows sketch the spin polarization. \( a \) is the lattice constant.
points emerge again. The Fermi arcs have a reversed spin polarization, owing to the change of sign of the SOC. Eventually, at $x \approx 0.24$, the pairs of Weyl points recombine at the mirror lines and the system enters the strong TI phase for $x > 0.24$.

It turned out that the Fermi arcs connect different pairs of Weyl points, depending on the surface terminations (Fig. 5). The Fermi arcs are reminiscent of Dirac strings that connect oppositely charged Dirac monopoles; this picture is supported by the spin polarization that follows a Fermi arc’s line (arrows in Fig. 5).

Concluding remarks.—Strained HgTe$_{\delta}$S$_{1-\delta}$ alloys show a surprisingly rich topological phase diagram: besides a reversal of the spin chirality in the strong topological insulator phase, we predict two semimetallic topological insulators, namely, a Dirac semimetal and a Weyl semimetal. These findings call for experimental verification, for example, by spin- and angle-resolved photoelectron spectroscopy [54,55]. In transport measurements, all the phases would have different signatures. In Weyl semimetals, the surface contribution should be highly anisotropic because of the shape of the Fermi arcs, which could be detected by surface sensitive measurements. In contrast, the anisotropy should nearly vanish in topological Dirac semimetals, since the Fermi lines are closed. Finally, for the STI phases, the bulk contribution should vanish because of the insulating bulk nature. This would not be the case in the semimetallic phases.

Positive or negative strain may be introduced by appropriate substrates, as is successfully done for HgTe sandwiched between CdTe (Ref. [14]). With the assumption that the lattice constant of HgTe$_{\delta}$S$_{1-\delta}$ varies linearly with $x$, we propose GaSb with a lattice constant of 6.1 Å [56] as a substrate for the Weyl semimetal phase. To achieve the topological Dirac semimetal phase, we propose Cd$_{0.7}$Zn$_{0.3}$Te (lattice constant 6.364 Å [57]) as a suitable substrate. Figure S3 of the Supplemental Material [58] provides in-plane lattice constants for the entire topological phase diagram.

In our work, the alloy was simulated mainly by the virtual crystal approximation. A big advantage is the possibility of calculating the topological invariants from eigenstates and eigenvalues combined with the fact that we do not need to construct large supercells. In the DFT approach, different methods for simulating alloys are possible. One is to construct a supercell, where the atoms of compound $A$ are exchanged by compound $B$ according to the given concentration. The system is then lattice periodic. The biggest disadvantage of this approach is the need for large supercells, which can become computationally demanding. It was used to calculate, e.g., the properties of Hg$_{0.7}$Zn$_{0.3}$Te [15], which is similar to the system we investigate. Another possibility is the CPA. Since this method is based on a Green function approach, it is well suited for our Korringa-Kohn-Rostoker package.

The next step would be to calculate the properties of alloys using the CPA directly within DFT.

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[18] It has to be noted that HgS has also been found to be a normal insulator with negative SOC [19].
Details of the computations are provided in the Supplemental Material [58], which includes Refs. [27–33].


