Ferroelectric Order Control of the Dirac-Semimetal Phase in GeTe-Sb$_2$Te$_3$ Superlattices

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The quasibinary GeTe-Sb$_2$Te$_3$ system (primarily the Ge$_2$Sb$_2$Te$_5$ composition, or GST225) has been long used in optical memory devices such as re-writable DVD-RAM and are also a leading candidate for the nonvolatile electronic memory known as phase-change random-access memory (PC-RAM); last year Samsung and Micron have started shipping these devices into the market. The basis of the phase-change storage is a large property contrast between the crystalline and amorphous phases; the idea dates back to the 1960s.[1] In a PC-RAM device, when a voltage exceeding a certain value (a threshold voltage) is applied to the high-resistivity amorphous phase, the material switches into the low-resistivity crystalline (SET) phase. The process can be reversed (RESET) by applying another pulse, of appropriately chosen amplitude and duration, that reverts the structure to the amorphous phase.

Non-volatile memory devices are currently key elements of various electronics and portable systems (digital camera, solid state disks, smartphones, computers, e-books, tablets, etc.) and their market has been increasing exponentially over the last decade. One important aspect is to develop new memory storage concepts and devices that can integrate multiple functionalities. It was recently found by some of the present authors[2] that when GeTe and Sb$_2$Te$_3$ components are spatially separated in forms of nm-thick layers to form a superlattice (interfacial phase-change memory, or iPCM), the energy efficiency of devices increases by orders of magnitude, the switching functionality of GeTe down to the nanometer scale. When a voltage exceeding a certain value (a threshold voltage) is applied to the high-resistivity amorphous phase, the material switches into the low-resistivity crystalline (SET) phase. The process can be reversed (RESET) by applying another pulse, of appropriately chosen amplitude and duration, that reverts the structure to the amorphous phase.

It is instructive to note that both individual constituents of iPCM possess rather specific properties. Sb$_2$Te$_3$ is one of the best known examples of three-dimensional (3D) topological insulators (TI).[3] GeTe, on the other hand, is the simplest known ferroelectric material with just two atoms in the primitive cell.[4] This conclusion was primarily reached based on structural studies;[5] the main challenge in direct studies of ferroelectricity in GeTe by conventional electrical spectroscopic techniques, such as hysteresis loop and transient current measurements, is its high conductivity: free charge carriers screen the applied electric field inhibiting polarization reversal and result in high dielectric loss. Ferroelectric order has been demonstrated for nm-sized nanocrystals,[6,7] and ferroelectric switching in GeTe has been observed experimentally in polycrystalline samples using piezoresponse force microscopy (PFM) and capacitance measurements.[8] Furthermore, stable ferroelectric switching has been recently observed in epitaxial GeTe films,[9] which provides tangible reasons to expect the scaling of the ferroelectric switching functionality of GeTe down to the nanometer scale. It is thus extremely interesting to combine these two materials in a superlattice and see how the ferroelectric order in the GeTe layer effects the TI properties of the overall structure.

In this work, we first study various possible stacking sequences of iPCM using density-functional theory and molecular dynamics. We demonstrate that one of the possible sequences forms the Dirac semimetal phase. We further demonstrate that upon application of an external electrical field the Dirac semimetal phase with inversion symmetry reversibly changes into a Rashba-split phase with ferroelectric ordering within the GeTe block, with a concomitant change in properties. For experimental verification of the simulational results, we have used iPCM superlattices with the $(\text{GeTe})_{10}(\text{Sb}_2\text{Te}_3)_{10}$ structure; the observed I–V characteristics are in line with the theoretical predictions. Finally, we discuss the relationship between these structural changes and the mechanism of data storage in iPCM and propose novel possibilities of using iPCM in devices with multiple functionalities.

The electronic band structures of iPCM films were simulated using two ab-initio simulation codes: CASTEP and WIEN2K. iPCM models were first relaxed using the plane wave code CASTEP with a GGA exchange correlation term as constructed by Perdew-Burke-Ernzerhof (PBE). A $7 \times 7 \times 1$ Monkhorst-Pack grid was used for integration and ultra-soft pseudopotentials were used with a cutoff energy of 230 eV.[10,11] In ab-initio molecular dynamics, NPT ensemble was used at 0 GPa. A cut-off energy of 170 eV was used. After 0 K Broyden geometrical optimization, the models were transferred to WIEN2K[12] to include spin-orbit coupling effects. Wien2K is an all electron code that uses a linearized augmented plane wave + local orbital (LAPW+lo) basis within density-functional theory using the PBE exchange-correlation. The Monkhorst-Pack grid of $7 \times 7 \times 1$ was used for integrations.
in the Brillouin zone and a $R_{\text{K}}_{\text{max}} = 7.0$ value was used for the plane wave component of the plane-wave basis used between augmentation spheres.

iPCM has the structure of a short-period superlattice, $[(\text{GeTe})_l(\text{Sb}_2\text{Te}_3)_m]_n$ (where $l$, $m$, and $n$ are integers), sharing a common growth axis: the $<111>$ direction of the rhombohedral GeTe layer and the $<111>$ axis of the rhombohedral (A7) $\text{Sb}_2\text{Te}_3$ layer are parallel to each other and normal to the substrate surface. Importantly, even sputtered iPCM devices exhibit a strong preferred $<111>$ growth direction and exhibit high quality interfaces due to the good lattice parameter match between GeTe and $\text{Sb}_2\text{Te}_3$.[2]

The structure of bulk $\text{Sb}_2\text{Te}_3$ can be described by a sequence of Te-Sb-Te-Sb quintuple layers (QLs) that possess spatial inversion symmetry.[3,13] Interatomic interactions within the QLs are covalent-like, with atoms in the interior of the QL forming multicenter bonds similar to the 3-center-4 electron bonds described earlier for Ge-Sb alloys.[14] In such bonding configurations, the central atom shares its electrons between the two bonds, i.e., the bonding can be described as resonant[15–17]Such bonds are usually softer compared to usual two-center covalent bonds between similar atoms. In cases when bond energy (length) asymmetry exists such as in Peierls-distorted structures, the shorter bond becomes even shorter with increasing temperature while the longer one elongates further.[18,19] The QLs are held together by much weaker forces, often referred to as being of van-der-Waals type.

Bulk GeTe has a rhombohedral (alternatively described as a distorted rocksalt) structure with a strong bonding energy hierarchy between the shorter and longer bonds,[18] which allows one to also consider its structure as layered, namely, covalently bonded buckled GeTe layers with much weaker interaction between the layers, similar to the case of $\text{Sb}_2\text{Te}_3$. Since Ge and Te atoms in the structure have different charges, there is a dipole moment oriented along the $<111>$ direction, i.e., perpendicular to the buckled GeTe layers. With the average composition of the superlattice being $\text{Ge}_2\text{Sb}_2\text{Te}_3$, and the thickness of the individual layers on the order of 1 nm, the structure can be imagined as an alternating sequence of two buckled GeTe layers and a single QL of $\text{Sb}_2\text{Te}_3$. How is the structure of these building blocks affected by making a GeTe-$\text{Sb}_2\text{Te}_3$ superlattice?

Experimental determination of layer sequences iPCM structures obtained experimentally is complicated by several circumstances, such as the extreme thinness of the layers, rather similar interlayer distances in all structures and structural imperfections arising from the use of sputtering technique (no high-quality epitaxial samples exist at present despite intense effort in this direction).[20] We have thus adopted a calculational approach, i.e., to compare the stability of the possible phases considered below.

First, we assume that the structure of the $\text{Sb}_2\text{Te}_3$ layer is the same as that of a QL in the bulk phase; a reasonable assumption considering the facts that this structure is stable, the thickness of the $\text{Sb}_2\text{Te}_3$ layers in iPCM is equal to that of a QL in the bulk phase within experimental accuracy, and the growth process of iPCM starts with a $\text{Sb}_2\text{Te}_3$ layer. We now turn to the structure of the GeTe block. The first possibility is that its structure is the same as that of the bulk phase, i.e., two neighbouring buckled GeTe layers with the same orientation of the dipole moments (the ferroelectric configuration). This structure is shown in Figure 1 in two different projections. The black lines indicate the unit cell. In what follows, we shall refer to this configuration as the ferroelectric GeTe phase and in subsequent figures we adopt the projection shown in the right panel.

At the same time, since GeTe is sandwiched by two identical QLs of $\text{Sb}_2\text{Te}_3$, i.e., the structure possesses mirror symmetry, an antiferroelectric configuration may also be possible. For the latter, two stacking possibilities exist, with either Ge or Te atoms located inside the GeTe block. The symmetric structure with the GeTe-TeGe layer sequence sandwiched by $\text{Sb}_2\text{Te}_3$ QLs is known in the literature as the Petrov sequence.[21] which was proposed to describe the stable trigonal phase of GST225. For obvious reasons, we shall refer to the other sequence, namely with the TeGe-GeTe order, as the inverted Petrov phase.

Figure 1. The structure of iPCM with ferroelectric GeTe order within the GeTe block shown in two different projections. Ge atoms are marked in green, Sb atoms in dark magenta, and Te atoms in orange and the black lines indicate the unit cell. In the following figures, we adopt the projection shown in the right panel.
in dark magenta and in Table 1 we summarise the results of ab-initio structure relaxation using DFT with and without spin-orbit coupling (SOC) for these structures. In addition, molecular dynamics simulations at the film growth temperatures (up to 600 K) have also been performed to investigate the effect of the growth temperature.

One can see from the Table that while at lower temperatures the Kooi phase has the lowest energy (and all models have rather small energy differences), in agreement with literature, at elevated (growth) temperatures of ca. 500 K, the Petrov and Kooi phases are energetically unfavorable and the inverted Petrov phase and the ferroelectric GeTe phase become energetically most favorable. The energy difference between the Kooi and Petrov phases compared to the inverted Petrov and ferroelectric GeTe phases further increases with increased temperature. We thus believe that the experimental iPCM phase is a mixture of the ferroelectric GeTe and inverted Petrov phases.

Finally, it should be mentioned that considering the structure of the stable trigonal phase as an alternative to the Petrov phase, another layer sequence with Ge and Sb layers interchanged has been argued by Kooi and Hosson to provide a better fit to the experimental results. The complete sequence in the Kooi phase is Te-Sb-Te-Ge-Te-Ge-Te-Sb-Te. The interaction between adjacent nonuple layers [Nonuple (Latin) = consisting of nine parts. We use this term analogously to the term “quintruple layer” used to describe the structure of Sb$_2$Te$_3$] is similar to that between quintuple layers in Sb$_2$Te$_3$. Two significant differences between the Kooi and Petrov phases should be noted. Firstly, while the GeTe block is located between the two Sb$_2$Te$_3$ QLs in the Petrov phase, it is incorporated into the Sb$_2$Te$_3$, forming nonuple layers in the Kooi phase. One should also note that the nonuple layer is terminated by Te atoms, similar to the case of Sb$_2$Te$_3$.

These four structures are compared in Figure 2, where the Ge atoms are shown in green, Te atoms in orange and Sb atoms in dark magenta.
In Figure 2, the bulk band structures for these four phases are shown. It is interesting to note that the inverted Petrov phase possesses bands that look very similar to the Dirac surface states of a 3D-TI with an important difference that these are bulk states. This result can be easily understood within the following arguments. The present system is a superlattice between TI (Sb$_2$Te$_3$) and a normal insulator (NI) (GeTe). On each side of a TI layer there is a Dirac-cone surface (interface) state. The surface Dirac cones can be classified in terms of spin chirality. We shall refer to the case with the upper cone having clockwise chirality as positive (+) helicity, and the one with the upper cone having counterclockwise chirality as negative (-) helicity. The “upper cone” here means the Dirac cone located above the Dirac point. The Dirac cones on the opposite surfaces (top/bottom) of TI slabs have opposite chiralities.[24] For very thin TI slabs they hybridize through the TI giving rise to gap opening.[25] In the present case, additional hybridization through the GeTe layers is present, which, as we show below, can significantly modify the electronic states. A similar idea has been explored from a different perspective.[26]

The present system can be described by the following effective Hamiltonian:[27,28]

$$H = \sum_{k} \sum_{i,j} \left[ v_F \tau^z (\sigma^i k_p - \sigma^i k_d) \delta_{ij} + \Delta \tau^z \delta_{ij} + \Delta_N \tau^z \delta_{ij+1} \right] c^\dagger_{i,k} c_{j,k}$$

(1)

where $\sigma^i$ and $\tau^i$ denote the Pauli matrices acting onto the spin and the surface index (top/bottom), respectively, and $v_F$ is a Fermi velocity. $|\Delta_T|$ and $|\Delta_N|$ represent hybridization of surface states through the same TI slab and the neighboring TIs, respectively. $\Delta$ is the annihilation operator of electrons on the i-th TI layer with in-plane wavevector $k$. We consider the case where the helicities of the Dirac cones are positive (+) for the top surface and negative (-) for the bottom surface of each TI layer, which is typical for the topological insulators found so far.[29] Let us call this a “+/-” sequence. The energy eigenvalues for this case are:[27]

$$E = \pm \sqrt{v_F^2 k^2 + \Delta^2},$$

(2)

where $\Delta^2 = \Delta_T^2 + \Delta_N^2 + 2\Delta_T \Delta_N \cos k_d d$, where $d$ is the sum of thicknesses of the TI Sb$_2$Te$_3$ layer and the NI GeTe layer.

According to Burkov and Balents[27] the band structure for the superlattice depends on the relative magnitudes of $|\Delta_T|$ and $|\Delta_N|$: The cases $|\Delta_T| > |\Delta_N|$ and $|\Delta_T| < |\Delta_N|$ correspond to the TI phase and NI phase, respectively. It can be intuitively understood as follows. Both TI and NI are gapped in the bulk. If $|\Delta_T| > |\Delta_N|$, the surface states on the two surfaces of the same TIs hybridize strongly and a band gap develops (Figure 3(a)). On the other hand, if $|\Delta_T| < |\Delta_N|$, the surface states on the surfaces of the neighboring TIs hybridize strongly and the phase becomes gapped. Eventually only two surface states on the outermost surfaces can be regarded as surface states of the whole superlattice (Figure 3(b)). As a result, the material is a 3D TI. Because the TI and NI are different phases, the bulk gap must close in between the two phases.

If, on the other hand, $|\Delta_T| > |\Delta_N|$, $\Delta^2$ in Equation (2) can become zero, the band gap closes and a bulk Dirac cone with linear dispersion is formed; this phase has been called a Dirac semimetal[27,30,31] For the particular case when $\Delta_T / \Delta_N = -1$, the Dirac point is formed at the $\Gamma$ point. We note that the signs of $\Delta_T$ and $\Delta_N$ can be the same or the opposite, depending on the material and the thicknesses of the TI and NI. For example, the surface-state gap in a thin film of Bi$_2$Se$_3$ has been calculated and it was found that the gap is proportional to $|\Delta_T|$ and the oscillation of the gap corresponds to the alternating sign of $\Delta_T$ in a thin film.[32]

Figure 2 demonstrates that of the four studied models, the inverted Petrov phase phase corresponds to the Dirac semimetal. The fact that the Dirac cones in certain sequences in iPCM correspond to the bulk phase has a great advantage over the TI surface states from an application perspective. Considering the overall similarity in the stacking sequence, one might also expect the Kooi phase to be a Dirac semimetal, however, because the GeTe block in the Kooi phase is located inside the Sb$_2$Te$_3$ block, the NI component is effectively removed, only hybridization across the TI phase remains, excluding the possibility of the formation of a Dirac semimetal phase.

According to the above discussion, the topological properties of a GeTe-Sb$_2$Te$_3$ superlattice can be tuned by an appropriate choice of thicknesses of individual blocks. While, as demonstrated above, the inverted Petrov phase is a Dirac semimetal, in similar structures with Sb$_2$Te$_3$ blocks consisting of a thicker GeTe block (four buckled layers in antiferroelectric arrangement) or a thicker Sb$_2$Te$_3$ block (two QLs), the Dirac semimetals states disappear (Figure 4), in agreement with the above arguments. In the latter case, the energy dispersion remains almost linear but the gap opens. Below, we shall demonstrate that a transition to the gapped state can also be induced by changing the ferroelectric order of GeTe preserving its nominal thickness; the latter being a much more appealing option from an application perspective.
The GeTe block with ferroelectric order is terminated by a Te plane at one interface and by a Ge plane at the other one (Figure 5, central panel). The Ge atoms are attracted towards the Te atoms terminating the neighbouring QL (due to the Coulomb interaction assisted by the applied external field) to establish Te–Ge bonds similar to those that exists the bulk phase of GST. The resulting asymmetry in the structure with the GeTe block located significantly closer to one of the adjacent QLs is evident from the DFT simulations result shown in the figure. It is also interesting to note that the longer Te–Te distance between the GeTe and Sb₂Te₃ blocks is similar to that for the second-nearest distance in GST, i.e., a real vacancy layer is in principle generated.

Within the physics of topological insulators, when an electrical field is applied perpendicular to the superlattice, spatial inversion symmetry is broken. In this case, the band structure is deformed by Rashba splitting, a result in agreement with the recent report of a giant Rashba effect induced by an electrical field in bulk GeTe. The resulting change in the electronic structure modifies the values of |Δₜ| and |Δₙ|, even when the overall structure of the phases remains the same. In other words, the external electric field breaks the balance between |Δₜ| and |Δₙ|, and the system becomes either a TI or a NI phase, both possessing a bulk energy gap. The field-induced destruction of the Dirac semimetal phase can be either transient, if the initial structure is restored after the field is turned off.

Indeed, if we apply an external electrical field to the inverted Petrov phase perpendicular to iPCM interfaces, current starts to flow across the structure, generating a certain amount of Joule heat, which facilitates atomic motion. As the field is increased, the antiferroelectric arrangement of the GeTe dual layers (usually energetically unfavourable) switches to the ferroelectric arrangement.

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off, or permanent, if a new structure is created, as in the case of ferroelectric switching of GeTe.

The structural transformation under the applied electrical field may in principle continue. Indeed, as a result of field-generated asymmetry caused by the established ferroelectric order in GeTe, the Sb₂Te₃ QL per se also loses spatial inversion symmetry with the Te-Sb distance for the outer layers located further from the GeTe block becoming the shortest. At the same time, the next Sb-Te distance becomes the longest as demonstrated by the DFT simulation results. Shown in Figure 5 (central panel) are the Sb-Te interatomic distances within the QL after the establishment of ferroelectric order in the GeTe phase. The numbers in the left panel show the same distances in the starting inverted Petrov phase. In addition to one Sb-Te interlayer distance (marked Δ in the figure) becoming longer (and the asymmetry is expected to increase with temperature) \(^{18,19}\) the forces acting on differently charged Sb and Te species due to the applied field further increase this (and only this) particular Sb-Te interplane distance. This is schematically illustrated in Figure 5 (central panel), where forces acting on Sb and Te atoms along the longer interplane distances are shown. At a certain combination of electrical field and temperature, the Sb atoms can flip into the vacancy layer, similar to ferroelectric switching of the GeTe block, generating a new (3.60 Å) Te/Te interface. The resulting layer sequence is the same as in the Kooi phase (Figure 5, right).

We now proceed to experimental verification of the proposed process. While direct evidence of the change in ferroelectric order in GeTe blocks is not available, there is indirect supporting evidence. First of all, the proposed transition from the Dirac semimetal phase with the inverted Petrov layer sequence into a gapped phase should manifest itself by a specific change in the resistivity. Namely, as Ge atoms are displaced and gradually flip into the ferroelectric arrangement, the \(|\Delta_F| = |\Delta_N|\) balance is broken and a gap opens, making the structure more resistive. Experimentally, under an applied electrical field, one should thus observe a gradual increase in resistivity followed by an abrupt drop at the phase transition. This prediction has been well reproduced experimentally using an iPCM-based device (Figure 6), where strongly non-Ohmic behavior has been observed. We further argue that an abrupt drop in resistivity associated with a crystal-crystal transition \(^{22}\) is likely to be due to the change in ferroelectric order. A crucial experiment would be to observe a change in ferroelectric order in iPCM structures using PFM.

Finally we would like to note that for both TI materials, such as Sb₂Te₃, characterized by interfacial spin currents, and for GeTe that was predicted to exhibit electrical field-induced Rashba splitting \(^{33}\) an external magnetic field can modify their properties. It is thus proposed that GeTe-Sb₂Te₃ superlattices (iPCM) may possess an interesting response to an applied magnetic field in combination with the external electrical field. Actually, some of the present authors have recently reported that iPCM devices without and with applied magnetic field exhibit markedly different switching behavior. \(^{14}\) Further studies may open previously unexplored possibilities of using iPCM for spintronic applications and are currently underway.

In conclusion, using DFT simulations we have shown that GeTe-Sb₂Te₃ superlattices with the appropriately chosen thicknesses of individual blocks may form a Dirac semimetal phase. An external electrical field can disrupt this phase by changing the ferroelectric order in the GeTe block and subsequently modifying the hybridization constants between the neighboring Dirac cones. The experimentally observed strong non-Ohmic behavior in iPCM devices followed by an abrupt drop in resistance associated with a crystal-crystal transition provides indirect support of the proposed model. In addition to demonstrating that GeTe-Sb₂Te₃ superlattices provide an interesting platform...
to investigate properties of topological insulators, such as the hybridization of the Dirac states, our results also provide new insights into the phase-change process in interfacial structures of iPCM and offer novel possibilities for designing phase-change memory devices with multiple functionalities. A part of this work was funded by JSPS though the FIRST Program initiated by CSTP.

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