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Ferromagnetic half-metallicity in half-Heusler AuMnSn:Te Alloy

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Abstract

Identifying a strategy for beneficial band-engineering is vital for the optimization of spintronic materials. In this study, we demonstrate the beneficial effects of electron doping on ferromagnetic (FM) half-Heusler AuMnSn\(_{1-x}\)Te\(_{x}\) (0 \(<\ x\ <\ 1\)) using state-of-the-art first-principles methods. By partially substituting the Sn site with a more ionic Te dopant, we predict a FM metal to a FM-half-metallic semiconductor (FM-HMS) transition at Te doping concentration \(x\ \sim\ 0.125\). The FM-HMS is observed to be stable over a wide range of lattice parameters up to a pressure of \(\sim\ 24.91\) GPa. We determined the magnetic phase diagram that shows a phase transition from FM state at low Te concentration dominated by the FM Ruderman–Kittel–Kasuya–Yosida-type exchange to antiferromagnetic state at high Te concentration mediated by the superexchange coupling mechanism. Using the Heisenberg exchange model, we predict a rather high Curie temperature of \(T_C\ \sim\ 725\) K that increased with increasing pressure. Our results demonstrate a robust scheme for engineering ferromagnetic half-metallicity in the half-Heusler alloy that is promising for spintronic applications.

1. Introduction

The search for ferromagnetic and stable half-metallic ferromagnetic semiconductors remains a high priority in designing next-generation electronic devices [1–3]. The use of the spin degree of freedom instead of or in addition to the charge degree of freedom (generally known as spintronics) is promising for efficient information processing and storage [4–9]. At present, ferromagnetic metals are still the most commonly used materials in spintronic devices. However, metal-based spintronic devices have a disadvantage when combined with semiconductors; a metal-semiconductor junction is prone to large Schottky barrier, which diminishes spin lifetime [10]. To increase the spin injection at the ferromagnetic metal-semiconductor interface, other materials such as ferromagnetic oxides, dilute magnet or magnetic semiconductors, and Heusler alloys [11–15] are being explored. However, most of these materials are characterized by low Curie temperature (\(T_C\)) [13, 16], high spin polarization only at low temperature [6, 17–20], and antiferromagnetic interaction that strongly suppress the \(T_C\) [21, 22].

Another class of materials that are promising for novel applications is the half-Heusler (HH) compounds. They exhibit one of the broadest set of exotic properties such as magneto-optical Kerr effect [23, 24] and as electrode materials in magnetic tunnel junctions [25]. These properties are promising for erasable data-storage technology and other diverse applications [26–28]. AuMn-based HH compounds [23, 24, 29–38] have been intensively studied, but most studies seem to posit an equiatomic composition. Some earlier works on HH AuMnSn and AuMnSb reported the deviation from equiatomic, i.e., 1:1:1 composition at \(\sim\ 400\ ^\circ\) C [29–31, 33, 36]. Crystal-orbital–Hamiltonian population reveal that neither AuMnSn nor AuMnSb displays an optimized bonding configuration in the equiatomic composition. The optimum bonding is achieved when all the bonding states are occupied, and the antibonding states are unoccupied [39].
Half-Heusler AuMnSn is a ferromagnetic metal exhibiting cubic Cl\textsuperscript{2} crystal structure (space group F\textsuperscript{4} 3 m, no. 216) (see, figure 1 (a)). The unit cell is made of three inter-penetrating face-centered-cubic sublattices with the Wyckoff positions as indicated between parentheses: Au: 4c \(\left(\frac{1}{2}, \frac{1}{2}, \frac{1}{2}\right)\), Mn: 4b \(\left(\frac{1}{2}, \frac{1}{2}, \frac{1}{2}\right)\), and Sn: 4a \(\left(0, 0, 0\right)\) [27]. To investigate the electronic and related properties of the AuMnSn-based half-Heusler compound, we performed first-principles calculations based on density functional theory (DFT) [41, 42] as implemented in the Quantum Espresso code [43]. The calculations were performed using the revised Perdew–Burke–Ernzerhof [44] exchange correlation functional that improves equilibrium properties of densely packed solids and their surfaces. We used a planewave basis sets with kinetic energy cutoff of 60 and 480 Ry for the wavefunctions and charge density, respectively, and a 12 \(\times\) 12 \(\times\) 12 Monkhorst–Packscheme [45] to represent the reciprocal space. Scalar relativistic ultrasoft pseudo-potentials [46] were used to describe the interaction between ions and valence electrons with Au: 6s, 5d; Sn: 5s, 5p, 4d; Mn: 3s, 4s, 3p, 3d; and Te 5s, 5p, 4d are considered as valence orbitals.

To model the defect structure, we constructed various supercell sizes large enough for the various Te defect concentrations in AuMnSn\textsubscript{1\textendash}xTe\textsubscript{x} (see, figure 1 (b) for \(x = 0.125\)). Pristine AuMnSn is known to exhibit a FM ground state and exhibits three crystalline forms: \(\alpha\), \(\beta\), and \(\gamma\) phases [29, 36]. To confirm the ground state and crystalline phase in our calculations, we carried out total energy versus lattice constants calculations for the various magnetic configurations and free energy calculations at various pressures. We obtained a ferromagnetic ground state [see, Supplemental Materials (SM)\footnote{See Supplemental Material at [URL will be inserted by publisher]. We show the determination of the magnetic ground state of the AuMnSn parent materials. Also, the optimized doping concentration for the transition from FM to FM-HM occurs around Te doping concentration of \(x = 0.125\). We show the electronic structure of other Te doping concentrations around the optimal one. We further demonstrate that the FM-HMS is stable against basis set with the all-electron method as implemented in Wien2k electronic structure code.}] available online at stacks.iop.org/MRX/7/076519/mmmedia]. Using the ferromagnetic ground state, we further

2. Computational details

Band- and defect-engineering are essential techniques to systematically control carriers in materials and achieve optimized bonding. In this study, we explore the effect of electron doping on the structural, mechanical, electronic and magnetic properties of AuMnSn\textsubscript{1\textendash}xTe\textsubscript{x} (0 \(\leq\) x \(\leq\) 1) using first-principles methods. We determine the magnetic phase diagram of our system and predict a ferromagnetic (FM) to antiferromagnetic (AFM) phase transition at \(\approx 0.46\). We attribute the observed magnetic phase transition to the competition between the ferromagnetic Ruderman–Kittel–Kasuya–Yosida (RKKY)-type exchange and antiferromagnetic superexchange [40]. We predict a robust ferromagnetic half-metallicity at \(x = 0.125\). The induced ferromagnetic half-metallicity is demonstrated to be stable over a wide range of lattice constants and exhibits a rather high Curie temperature of \(\sim 725 \text{K}\), which increased with increasing pressure. The predicted properties could be useful in spintronics and other novel optoelectronic applications.

Figure 1. (a) Primitive unit cell of half-Heusler AuMnSn (space group F\textsuperscript{4} 3m) (b) AuMnSn\textsubscript{0.875}Te\textsubscript{0.125} in the Cl\textsuperscript{2} structure (space group F\textsuperscript{4} 3m) with Sn/Te, Au and Mn at (0, 0, 0), \(\left(\frac{1}{2}, \frac{1}{2}, \frac{1}{2}\right)\) and \(\left(\frac{1}{2}, \frac{1}{2}, \frac{1}{2}\right)\) positions, respectively.

![Figure 1](image-url)
confirmed that the $\gamma$-phase is the most energetically stable phase; (see footnote 5) the $\gamma$-phase has been used in subsequent calculations presented below. The observed ferromagnetic $\gamma$-phase ground state is in agreement with experiments [29, 36]. The equilibrium lattice constant of AuMnSn$_{1-x}$Te$_x$ is obtained by computing the total energy at the various lattice constants around the experimental lattice constants of AuMnSn and fitting the calculated lattice constants and the total energy into the Murnaghan equation of state [47]. The position of all the atoms in the superstructure are relaxed using the Broyden-Fletcher-Goldfarb-Shanno method [48, 49] until the energy (charge) is converged to within $10^{-3}$ ($10^{-5}$) Ry and the Hellmann-Feynman forces dropped to $\sim 10^{-3}$ Ry/Å. We note that the optimized AuMnSn$_{1-x}$Te$_x$ structure retains the Cm$\bar{3}$m symmetry of the parent material. Using the predicted equilibrium lattice constant of AuMnSn$_{1-x}$Te$_x$ (see table 1), we proceeded to carry out self-consistent spin-polarized calculations.

An important parameter in determining the applicability of any material as a HMF in spintronic-based devices is the ease with which it can be spin polarized. It is beneficial if the polarization of the electron spins in the vicinity of the Fermi level $E_F$ is large. In electronic structure calculations, the spin polarization $P$ is calculated as [50] $P = (N_{\uparrow} - N_{\downarrow}) / (N_{\uparrow} + N_{\downarrow}) \times 100\%$, $N_{\uparrow\downarrow}$ denotes the density of states at the Fermi level. The exchange coupling between the magnetic moments is studied by mapping the energy differences of the various magnetic configurations to a Heisenberg Hamiltonian [51, 52] within the first and second-nearest-neighbour approximation.

### 3. Results and discussions

#### 3.1. Thermodynamic stability

The thermodynamic stability is determined by calculating the formation energy $E_{\text{form}}$ per formula unit (f.u.). The $E_{\text{form}}$ determines the energetic stability of a material and for our system, it is obtained as $E_{\text{form}} = E_{\text{tot}} - \sum_i c_i \mu_i$, where $i$ denotes different types of atoms present in the unit cell of the material with concentration $c_i$ and $\mu_i$ is the chemical potential approximated with the standard state (bulk) energy $E_i$ of the corresponding atom, $i$. As presented in table 1, both structures show negative formation energies, which implies an exothermic process, and it is a confirmation of the thermodynamic stability of the defect structures.

To establish the possibility of synthesizing the AuMnSn$_{0.875}$Te$_{0.125}$ alloy, we further calculate the defect formation as $E_d = E^{\text{Si}} - E^{\text{Ti}} - E^{\text{Si}} + E^{\text{Sn}}$, where $E^{\text{Si}}$ is the total energy of AuMnSn$_{0.875}$Te$_{0.125}$, $E^{\text{Si}}$ is the total energy of equivalent supercell size of AuMnSn, and $E^{\text{Te}} (E^{\text{Sn}})$ is the total energy per Te (Sn) atom in the bulk Triclinic (cubic) lattice. We obtained $E_d \approx 0.54$ eV; the low value favors the energetic formation of AuMnSn$_{0.875}$Te$_{0.125}$. To further confirm the stability of the $\gamma$-phase for the alloy composition, we calculate the free energy profile ($\Delta G = \Delta H - T\Delta S$, where $H, S,$ and $T$ is enthalpy, entropy, and temperature, respectively, for DFT, $T = 0$ K) as a function of pressure (figure 2). The $\gamma$-phase is confirmed to be the most energetically stable over a wide range of pressure up to $\sim 100$ (130) GPa where we predict a structural phase.

### Table 1. Elastic constants and mechanical properties of half-Heusler compounds AuMnSn$_{1-x}$Te$_x$. Herein, $a$, $E_{\text{form}}$, $B$, $Y$, $\nu$, $A$, and $B/G$ are the lattice constant, formation energy, bulk modulus, Young modulus, shear modulus, Poisson ratio, elastic anisotropy, and Pugh’s indicator, respectively. $P_f$ denotes pressure in GPa.

| Parameter | AuMnSn | AuMnSn$_{0.875}$Te$_{0.125}$ |
|-----------|--------|-----------------------------|
| $a$ (Å)   | —      | 6.323$^a$                   |
| $E_{\text{form}}$ (eV) | $-0.492$ | $-0.425$ |
| $C_{11}$ (GPa) | 103.1  | 115.2                      |
| $C_{12}$ (GPa) | 76.4   | 86.9                       |
| $C_{44}$ (GPa) | 34.2   | 36.1                       |
| $B$ (GPa)   | 85.3   | 96.4                       |
| $Y$ (GPa)   | 64.4   | 68.4                       |
| $G$ (GPa)   | 23.5   | 24.8                       |
| $\nu$       | 0.3730 | 0.3806                     |
| $A$         | 2.562  | 2.551                      |
| $B/G$       | 3.630  | 3.508                      |

$^a$ Reference [29] is experimental result at 150 K;

$^b$ Reference [35] is a theoretical result obtained with the Voigt-Reuss-Hill approximation.
transition from $\gamma$–to $\beta$–phase in AuMnSn$_{0.875}$Te$_{0.125}$ (AuMnSn). This transition pressure is quite high relative to the value of 24.91 GPa we considered in our modeling.

3.2. Elastic and mechanical properties

We studied the mechanical stability by calculating the elastic constants. Herein, we focus on the optimal Te doping concentration $x \approx 0.125$. In the absence of any external load, the elastic energy associated with the stress tensor $\sigma_{ij}$ due to a small strain $\epsilon_{ij} = \sum_{i=1}^{n} C_{ij} \epsilon_{ij}$ is expected to be positive definite [54]. For a cubic lattice, the elastic constants $C_{41}, C_{12}$, and $C_{44}$ are the most essential to establish the stability criteria [55]. We obtained the elastic tensor by inducing small but finite distortions on the lattice to generate series of stress-strain relationship following the approach described in [56] We present in table 1 the calculated elastic constants. Observe that $C_{11}$ is $\approx$ 3-times larger than $C_{44}$ in AuMnSn$_{1-x}$Te$_x$. This suggests that AuMnSn$_{1-x}$Te$_x$ offers a stronger resistance to deformation in one direction than to pure shear deformation. We observe that the resistance increased slightly with pressure in AuMnSn$_{1-x}$Te$_x$. As shown in table 1, the elastic constants are positive definite and satisfy the Born-Huang stability criteria ($C_{11} - C_{12} > 0$, ($C_{11} + 2C_{12} > 0$, and $C_{44} > 0$ [57, 58]; confirming the mechanical stability of AuMnSn$_{1-x}$Te$_x$.

We further obtained from the elastic constants calculations the bulk modulus $B$, Young’s modulus $Y$, and the shear modulus $G$, using the Voigt-Reuss-Hill approximation [59, 60]. The elastic moduli are obtained as $B = (C_{11} + 2C_{12})/3$, $G_Y = (C_{11} - C_{12} + 3C_{44})/5$, and $G_R = 5(C_{11} - C_{12})C_{44}/[4C_{44} + 3(C_{11} - C_{12})]$ . Using the above relations, we obtain $Y = 9BG/(3G + B)$ [61], the Poisson ratio $\nu = 3B - 2G/2(3B + G)$ [61], the Pugh’s indicator $B/G$ [62], and the elastic anisotropy factor $A = 2C_{44}/(C_{11} - C_{12})$ [63] as presented in table 1. Observe that AuMnSn$_{1-x}$Te$_x$ ($x = 0.125$) exhibited better elastic properties than AuMnSn. While the Poisson ratio remains almost constant for both systems even at higher pressure, the Young’s and the bulk modulus of AuMnSn$_{1-x}$Te$_x$ ($x = 0.125$) increased significantly at a pressure of 24.1 GPa. A material is ductile if $B/G > 1.75$ [64] and $\nu > 0.26$. As could be confirmed from table 1, AuMnSn$_{1-x}$Te$_x$ is ductile and the ductility increases with pressure. The shear anisotropy factor for an isotropic crystal is expected to be 1, and the anisotropy is reflected in the deviation from 1, i.e, $\lambda > 1$. Our results show that AuMnSn$_{1-x}$Te$_x$ is anisotropic. The result for AuMnSn is in agreement with the theoretical result of Amudhavalli et al [35]. We are not aware of any experimental data on the elastic and mechanical properties of HH AuMnSn-based alloys.

3.3. Electronic properties

We present in figures 3 and 4 the electronic properties of AuMnSn$_{1-x}$Te$_x$ obtained using spin-polarized DFT calculations. From the total density of states plotted around the Fermi level (figures 3(a)–(c)), we predict a $P$ of $\sim$28 and 100% for AuMnSn and AuMnSn$_{0.875}$Te$_{0.125}$, respectively. We observed that the spin polarization decreased below and above the optimal doping concentration. For example, the $P$ at $x = 0.0625$ and 0.1875 is 60 and 70%, respectively. The 100% spin polarization obtained at $x = 0.125$ persists and is stable up to a lattice constant of 6.244 Å, which corresponds to a pressure of $\sim$24.91 GPa. We also benchmarked the stability of the predicted properties using the all-electron Wien2k electronic structure code [65]. We obtain a 100% spin
polarization and a minority spin gap of $\sim 0.28$ eV. (see footnote 5) in good agreement with our reported results. We note that there is a finite density of states at Fermi level of AuMnSn in both the spin channels (figures 4(a) and (c)) confirming a FM metallic ground state and absence of half-metallicity. Focusing on the states around the Fermi level, the effect of Te doping is not a trivial shift of the band structure. In both spin channels, the Fermi level is shifted upward. Noticeably, in the minority spin channel (figure 4(d)), the doping process induced almost triple degenerate states at the conduction band minimum (i.e., the G point), which is nondegenerate in AuMnSn (figure 4(c)).

To gain further insights into the electronic properties of HH AuMnSn alloy, we explore the projected density of states (figures 3(d)–(i)). The electronic structure of AuMnSn$_{1-x}$Te$_x$ is characterized by a large exchange splitting of the Mn $d$ states. The $d$ states of the minority spin channel of Mn are generally unoccupied due to strong exchange splitting of Mn while that of Au are occupied. We attribute the minority spin gap to the bonding and antibonding Mn and Au $d$ states. For AuMnSn, the states around the Fermi level are predominantly due to the hybridization of the $e_g$ and $t_{2g}$ states of both Au and Mn, respectively. Specifically, those of the Mn $e_g$ and $t_{2g}$ states with a small contribution from Au $d$ states dominate the minority spin channel. Doping AuMnSn with Te implies introducing $s$ and the polarizing $p$ orbitals. As observed in figures 3(d)–(i), the $p$ states are extended and lower in energy than the $d$ states of Au and Mn, respectively. This could be interpreted as the increase in the occupation due to the injection of extra electrons into the material, which pushes the Fermi level to higher energy, thereby completely filling the minority band bonding orbital.

3.4. Magnetic properties

We explore the magnetic properties by calculating the spin magnetic moment, the exchange coupling parameters, and the Curie temperature. To expose the mechanism for the emergence of the ferromagnetic half-metallic semiconductor (FM-HMS), we explore in more details the electronic properties and atom-resolved spin magnetic moments for the FM configuration presented in table 2. One of the properties of half-Heusler materials is the deviation from the valence-electron content (VEC) [32, 33], which is the required number of valence electrons as prescribed by the Periodic table. The crystal-orbital-Hamiltonian-population analysis of Offernes and co-workers [32] proposed that the maximum occupation of the bonding states in AuMnSn exists at $\sim 0.01$ eV above the Fermi level; implying the addition of electrons up to the minority spin gap. The integration of the density of states suggests that the addition of 0.2 electrons can be contained in the bonding states, which implies that the VEC for optimal stability in AuMnSn is not 22 [e] but $\sim 22.2$ [e]. Hence, when the Sn site is partially substituted with a more ionic Te atom, it provides the additional 0.2 electrons required to achieve optimal VEC in AuMnSn. From our calculations, we predict that a Te doping concentration $x = 0.125$, i.e., AuMnSn$_{0.875}$Te$_{0.125}$, possess the optimal VEC which maximize the bonding interactions leading to the
We predict a direct energy bandgap $E_g \sim 0.30$ eV in the minority spin channel (figure 4(d)). We observe that the application of pressure up to 24.91 GPa has a net negligible effect. Interestingly, the FM-HMS is lost at below and above the optimal doping concentration (see, SM see footnote 5) supporting the assertion of the existence of optimal filling of the bonding orbitals required to achieve optimal VEC in AuMnSn.

We explore the origin of the FM-HMS behavior in AuMnSn$_{0.875}$Te$_{0.125}$ by studying the atomic resolved spin magnetic moment. The spin magnetic moments of Au and Sn in AuMnSn upon Te doping change from $-0.01$ and $-0.17 \mu_B$ to $0.04$ and $-0.11 \mu_B$, respectively, and the Te atom acquired a spin magnetic moment of $\sim 0.07 \mu_B$. Because of the almost filled electronic shells of these atoms, their contribution to the total spin magnetic moment is small. The Mn atom spin magnetic moment for AuMnSn is obtained as $4.05 \mu_B$; this implies that the five majority spin states are mostly occupied. However, upon Te doping, the extra electrons increase the hybridization of the Sn/Te with the Au and Mn-d bands, especially in the majority spin channel due to the more spread in space. The $p$ states push the Fermi level to higher energy as their occupation is increased because of their significant energy extension. Thus, the antiparallel spin magnetic moment coupling of Sn/Te to Mn atom in pristine AuMnSn is reduced upon Te doping while increasing the magnitude of the positive spin magnetic moment of Au and Mn atoms. Based on the Slater–Pauling (SP) rule [66], the total spin magnetic moment per unit formula $M_s$ of $4.0 \mu_B$/f.u and $4.25 \mu_B$/f.u is required for AuMnSn and AuMnSn$_{0.875}$Te$_{0.125}$, respectively to be a perfect half-metal ferromagnet. The SP rule [66] postulates that the total spin moment in $\mu_B$ of a perfect half-metallic ferromagnet is the number of valence electrons minus 18, i.e., $M_{SP}^s = N_v - 18$, where

![Figure 4. Spin resolved electronic band structures of AuMnSn (first column), AuMnSn$_{0.875}$Te$_{0.125}$ (second column). The upper panel is the majority spin band structure while the lower panel is the minority spin band structure. The Fermi energy is at zero energy represented by the dashed horizontal line.](image-url)
of 0 and 24.91 GPa, respectively, and above room temperature, which suggests that Te-doped AuMnSn could be a good material for spintronic-based device applications.

Our calculations show that the ferromagnetic exchange interaction in AuMnSn0.875Te0.125, we also predict rather large FM interactions, while negative ones correspond to AFM interactions. Assuming the second nearest-neighbour exchange coupling parameters, respectively, and $J_1$ and $J_2$ are the first and second nearest-neighbor exchange coupling parameters, respectively. Our calculations show that the ferromagnetic exchange interaction in AuMnSn1−$x$Te$_x$ compounds emanate mainly from an indirect exchange interaction mediated by the conduction electrons from the Au and/or Sn/Te atoms. Our observations are consistent with experiment and previous theoretical results [29, 69]. The calculated $T_C$ is 844 K for AuMnSn (table 2) is in good agreement with the experimental results [29, 31]. For AuMnSn$_{0.875}$Te$_{0.125}$, we also predict rather large $T_C$ ~ 725 K, which increased with increase in pressure (table 2). We note that the mean-field theory usually overestimates the Curie temperature compared to the experiment, mainly because it estimates the $T_C$ by averaging the magnon energies of the crystal lattice and thus neglecting its local variations. Nevertheless, the values of the $T_C$ we have obtained is comparable to the experiment and is well above room temperature, which suggests that Te-doped AuMnSn could be a good material for spintronic-based device applications.

### Table 2. Local spin magnetic moment and total spin magnetic moment per formula unit $M_s$ (in $\mu_B$) for pressure $P$, of 0 and 24.91 GPa, first and second nearest neighbour exchange coupling parameters $J_1$ and $J_2$ (in meV), Curie temperature (in K), for HH AuMnSn and AuMnSn$_{1-x}$Te$_x$ ($x = 0.125$).

| Parameter | AuMnSn | AuMnSn$_{1-x}$Te$_x$ |
|-----------|--------|----------------------|
| Present work | Other work | $P_1 = 0$ | $P_2 = 24.91$ |
| $M_{sa} (\mu_B)$ | $-0.01$ | $-0.022^a$ | $0.04$ | $0.05$ |
| $M_{sm} (\mu_B)$ | $4.23$ | $4.192^a$ | $4.27$ | $4.24$ |
| $M_{sn} (\mu_B)$ | $-0.17$ | $-0.155^a$ | $-0.11$ | $-0.10$ |
| $M_{s} (\mu_B)$ | $4.05$ | $4.089^a, 3.8^b$ | $4.25$ | $4.25$ |
| $J_1$ (meV) | $0.863$ | $0.769$ | $0.809$ |
| $J_2$ (meV) | $0.351$ | $0.248$ | $0.239$ |
| $T_C$ (K) | $843.74$ | $740^f, 625 \pm 100^f$ | $725.40$ | $754.62$ |

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N$_x$ is VEC. As presented in table 2, the $M_s$ at zero pressure for AuMnSn$_{0.875}$Te$_{0.125}$ is ~ 4.25 $\mu_B$ in basic agreement with the SP rule for a perfect half-metallic ferromagnetic material. The loss of the FM-HMS at higher Te doping concentration, i.e., beyond the optimal doping value could be attributed to the energetically preference of the electrons to occupy the spin minority states above the energy gap after reaching the optical VEC due to the large energy that will be required for any additional electron to occupy the Mn majority states beyond the optimal bonding states.

We estimate the strength of the magnetic interactions by calculating the interatomic exchange coupling constants between Mn-Mn atoms using the classical Heisenberg Hamiltonian [51, 52] $H = -2\sum_{i>j} J_{ij} S_i S_j$, where the localized spin $S = \frac{5}{2}$ is the total moment ($L = 0$ for half-filled shells) on Mn$^{2+}$ at site $\mathbf{r}_i$, and $J_{ij}$ is the exchange integral between the spins localized at sites $\mathbf{r}_i$ and $\mathbf{r}_j$. The exchange coupling parameters can be calculated by correlating the total energy of the ferromagnetic (FM), first antiferromagnetic (AFM 1), and second antiferromagnetic (AFM 2) configurations. In AFM 1, localized spins on Mn atoms are ferromagnetically aligned within the (001) planes which then alternate along the {011} direction. In AFM 2, spins are aligned in the (111) plane, which alternates along the {111} direction. The energies for the different configurations considered can be written as [67] $E_{FM} = E_0 + \zeta (-12I_1 - 6I_2)$; $E_{AFM_1} = E_0 + \zeta (4I_1 - 6I_2)$; and $E_{AFM_2} = E_0 + \zeta (6I_2)$, where $E_0$ is the energy of the independent spin configuration and $\zeta = S(S + 1)$. We have included $J_{ij}$ to $J_{ij}$; it drops fast as the Mn-Mn distance increases, e.g., $J_{ij} \sim 0.032$ meV. Positive values of the coupling parameters correspond to FM interactions, while negative ones correspond to AFM interactions. Assuming the second nearest neighbour approximation within the mean-field theory, the Curie temperature $T_C$ is then computed as $T_C = \frac{25S(S + 1)}{3\zeta} (Z_1 I_1 + Z_2 I_2)$ [68], where $Z_1$ and $Z_2$ are the number of first and second nearest neighbors respectively, and $I_1$ and $I_2$ are the first and second nearest-neighbor exchange coupling parameters, respectively. Our calculations show that the ferromagnetic exchange interaction in AuMnSn$_{1-x}$Te$_x$ compounds emanate mainly from an indirect exchange interaction mediated by the conduction electrons from the Au and/or Sn/Te atoms. Our observations are consistent with experiment and previous theoretical results [29, 69]. The calculated $T_C \sim 844$ K for AuMnSn (table 2) is in good agreement with the experimental results [29, 31]. For AuMnSn$_{0.875}$Te$_{0.125}$, we also predict rather large $T_C \sim 725$ K, which increased with increase in pressure (table 2). We note that the mean-field theory usually overestimates the Curie temperature compared to the experiment, mainly because it estimates the $T_C$ by averaging the magnon energies of the crystal lattice and thus neglecting its local variations. Nevertheless, the values of the $T_C$ we have obtained is comparable to the experiment and is well above room temperature, which suggests that Te-doped AuMnSn could be a good material for spintronic-based device applications.
3.5. Magnetic phase transition

As a function of increasing Te doping concentration, we observe a transition from ferromagnetic metal to ferromagnetic half-metallicity at $x \sim 0.125$ and then to antiferromagnetic phase at $x \gtrsim 0.46$. We broadly classify this process as FM to AFM transition. In order to understand the origin of the observed magnetic phase transition, we compute the total energies corresponding to the FM and AFM configurations of the Mn magnetic moments. We employed the same large unit cell for all the calculations. The magnetic phase diagram (figure 5) is obtained as the difference between the total energies of the AFM and FM magnetic configurations, i.e.,

$$\Delta E = E_{AFM} - E_{FM}$$

per unit cell. As shown in figure 5, the FM to AFM transition occurs at a critical Te concentration of $x \sim 0.46$. To the best of our knowledge, we are not aware of any experimental characterization of the magnetic phase diagram of AuMnSn$_{1-x}$Te$_x$. We note an earlier calculations by Özdoğan et al [34] on a related ternary system AuMnSn$_{1-x}$Sb$_x$ that predicted an FM to AFM transition at $x \sim 0.7$. We, however, remark that the magnetic phase transition depends sensitively on the lattice parameters since, e.g., pressure could push the Fermi level to higher energies leading to the increase in the superexchange interactions, which will support the AFM phase. While the FM-HMS behavior only emerges at $x \sim 0.125$, the ferromagnetic behavior is stable up to the critical point. We can qualitatively explain the FM to AFM magnetic transition in terms of the competition between the RKKY and antiferromagnetic superexchange [40]. As the Te concentration is increased, the Fermi level is pushed to higher energies, especially for the minority spin gap; this diminishes the strength of the exchange interactions. For Mn-based half-metallic Heusler compounds, the overall magnetic interactions strongly depend on the position of the Fermi level within the semiconducting spin channel [40]. As the Fermi level moves towards higher energy due to the increase in Te concentration, it enters into the conduction states of the minority spin channel at the transition point. The FM RKKY-like interactions between two neighboring Mn atoms are suppressed as the critical point is approached through the superexchange coupling of the Mn-Mn spin moments mediated through the Te atoms.

4. Conclusions

The magnetic and related properties of half-Heusler AuMnSn$_{1-x}$Te$_x$ compound has been investigated using first-principles techniques. We show that the partial substitution with an sp$^3$-electrons derived from Te atom on the Sn site induces a robust transition from a ferromagnetic metallic to ferromagnetic half-metallic semiconductor with 100% spin polarization at Te optimal doping concentration of $x \sim 0.125$. The induced FM-HMS is observed to be stable up to a hydrostatic pressure of 24.91 GPa. We determine the magnetic phase diagram, which showed an FM to AFM phase transition around $x \simeq 0.46$. Using a mean-field approach based on the Heisenberg Hamiltonian, we predict a large Curie temperature, which increased with increasing pressure.
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