Thermodynamical and topological properties of metastable Fe\textsubscript{3}Sn

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The Fe–Sn-based kagome compounds attract intensive attention due to its attractive topological transport and rich magnetic properties. Combining experimental data, first-principles calculations, and Calphad assessment, thermodynamic and topological transport properties of the Fe–Sn system were investigated. Density functional theory (DFT) calculations were performed to evaluate the intermetallics' finite-temperature heat capacity ($C_p$). A consistent thermodynamic assessment of the Fe–Sn phase diagram was achieved by using the experimental and DFT results, together with all available data from previous publications. Here, we report that the metastable phase Fe\textsubscript{3}Sn was introduced into the current metastable phase diagram, and corrected phase locations of Fe\textsubscript{5}Sn\textsubscript{3} and Fe\textsubscript{3}Sn\textsubscript{2} under the newly measured corrected temperature ranges. Furthermore, the anomalous Hall conductivity and anomalous Nernst conductivity of Fe\textsubscript{3}Sn were calculated, with magnetization directions and doping considered as perturbations to tune such transport properties. It was observed that the enhanced anomalous Hall and Nernst conductivities originate from the combination of nodal lines and small gap areas that can be tuned by doping Mn at Fe sites and varying magnetization direction.

**INTRODUCTION**

The kagome lattice is a 2D network of corner-sharing triangles that has been intensively investigated for over a hundred years. Due to its unusual geometry, it offers a playground for studying interesting physics, including frustrated, correlated\textsuperscript{1,2}, exotic topological quantum\textsuperscript{1–16}, topological Chern\textsuperscript{17} and Weyl semimetal\textsuperscript{13,18} phases, originating from the interplay between magnetism and electronic topology. In fact, the kagome lattice has been realized in several materials including metal stannides, germanides\textsuperscript{19,20} as well as $T_{n}X_{m}$ compounds with $T = Mn$, Fe, Co, X = Sn, Ge ($m:n = 3:1$, 3:2, 1:1)\textsuperscript{21}. Recent studies demonstrated that Fe–Sn-based kagome compounds exhibit interesting properties, such as large magnetic tunability\textsuperscript{1}. Furthermore, they can host Dirac fermions and flat bands, as found in Fe\textsubscript{3}Sn\textsubscript{2}\textsuperscript{22,23} and FeSn\textsubscript{2}\textsuperscript{24,25}. The existence of spin degenerate band touching points was linked to the generation of several interesting phenomena. Specifically, the anomalous Hall effect (AHE) results in a transverse spin-polarized charge current (charge current and spin current due to the imbalance of spin up and spin down electrons in ferromagnets) in response to a longitudinal charge current, in the absence of an external magnetic field\textsuperscript{25–29}. This applies also to its thermal counterpart, the anomalous Nernst effect (ANE), in which the external stimuli is replaced by a thermal gradient\textsuperscript{30} as well as the Seebeck effect\textsuperscript{31}.

Interestingly, the Fe–Sn-based intermetallic compounds not only exhibit attractive topological transport properties, but also show rich magnetic properties. In our previous studies\textsuperscript{32,33}, a DFT screening of the Fe–Sn phase diagram was used to identify Fe–Sn-based phases with the potential to be stabilized upon alloying, and their magnetization and magnetocrystalline anisotropy were evaluated. The results revealed that a strong anisotropy as observed in Fe\textsubscript{5}Sn may also be found in other Fe–Sn-based phases, having high potential to be used as hard magnetic materials. Meanwhile, we applied the reactive crucible melting (RCM) approach to the Fe–Sn binary system, and observed three metastable intermetallic compounds, namely Fe\textsubscript{3}Sn, Fe\textsubscript{5}Sn\textsubscript{3}, Fe\textsubscript{3}Sn\textsubscript{2}, which are ferromagnetic and exist between 873 K and 1173 K. We found that such metastable phases can be synthesized using the RCM method at specific temperature ranges. What’s more, phase diagram of the Fe–Sn system reported in the literature\textsuperscript{19,34,35} has mentioned that the Fe\textsubscript{3}Sn was considered to be a metastable phase, and presented the relevant so-called metastable composition range and phase relations. According to Fayyazi’s\textsuperscript{36} work, the reactive crucible reproduced the corresponding phase relations as in the bulk samples at 998 K (α-Fe, Fe\textsubscript{5}Sn\textsubscript{3}, FeSn, and Sn) and 1023 K (Fe\textsubscript{3}Sn\textsubscript{2}, Fe\textsubscript{3}Sn\textsubscript{2}, Fe\textsubscript{5}Sn\textsubscript{3}, and FeSn), of which Fe\textsubscript{3}Sn can only be stabilized between 1023–1098 K during a non-equilibrium process as a metastable phase but disappears at 1123 K due to the presence of Fe\textsubscript{3}Sn\textsubscript{2} phase. Accordingly, adding more details to the phase diagram of the metastable Fe\textsubscript{3}Sn phase, with the discovered temperature range based on the reported phase diagram is of great significance. Therefore, to further explore the interesting properties of metastable Fe–Sn phases, it is important to understand the phase diagram and thermodynamical properties of the Fe–Sn system.

In this work, we adopted our new measurements\textsuperscript{32,33} on the equilibria states of Fe\textsubscript{3}Sn, Fe\textsubscript{5}Sn\textsubscript{3}, Fe\textsubscript{3}Sn\textsubscript{2}, combined with the thermodynamic properties of such intermetallic phases obtained based on first-principles calculations. A consistent thermodynamic assessment of the Fe–Sn system was then developed based on all available experimental and first-principles results. Furthermore, the AHC and ANC of Fe\textsubscript{3}Sn were calculated and its dependence on the magnetization direction and doping were evaluated. We observed that there exist significant changes in AHC and ANC by...
tuning the Fermi energy via Mn-doping. Therefore, Fe$_3$Sn renders itself a promising candidate for new transverse thermoelectric devices with potential applications.

RESULTS AND DISCUSSION

Metastable phase diagram

Most end-members in the sublattice models are not stable and their thermodynamic data are impossible to be determined by experiments. First principles are hence performed to estimate the Gibbs energies of the compounds and end-members at finite temperatures. In order to benchmark the current DFT calculations, the calculated crystallographic information of phases in the binary Fe–Sn system are listed in Table 1, in comparison with the available experimental data. The calculated lattice parameters of the solid phases at 0 K are in good agreement with the experimental results at room temperature. As one can see, the differences between the theoretical and experimental lattice constants are within 0.5% for all the phases. Note that, in our earlier study, we showed, that the crystal structure of Fe$_2$Sn$_3$ synthesized by the equilibrated alloy method, is not of the typically assumed hexagonal Laves structure (as shown in Table 1). We rather observed superstructure reflections in the powder XRD spectra that could not be explained by the hexagonal structure and we assigned to a modulated orthorhombic unit cell with lattice parameters of $a = 4.221$ Å, $b = 7.322$ Å, $c = 5.252$ Å. More details and explanations can be found in the refs. Hence, we used this structure to do phonon calculations. Furthermore, the calculated phonon bands of such phases are shown in Fig. 1. To prove the validity of the calculations, as shown in Fig. 1, the phonon dispersion of BCC-Fe is compared with the experimental data, presenting good agreement. Therefore, it is expected that the thermodynamical properties of the Fe–Sn intermetallic phases can also be accurately obtained based on DFT calculations. As shown in Fig. 1, no imaginary phonon modes exist for all the compounds, indicating that all the intermetallics are dynamically stable. And the quasi-harmonic approximation (QHA) can be used to calculate the thermodynamic properties.

The thermodynamic properties at finite temperatures are evaluated based on the Gibbs free energies specified in Eq. (5). From the thermodynamical point of view, we can derive the magnetic contribution to the heat capacity following the theory of IHJ model and further improved version by Xiong:

$$C_{p\text{mag}} = R(\beta^2 + 1)c(\tau).$$

(1)

Figure 2 shows isobaric heat capacity obtained from our DFT calculations. It can be observed that the lattice vibrations dominate other contributions to the heat capacity. Interestingly, the correction made by adding electronic and magnetic heat capacities shifted the result toward bigger values and after that calculations show an excellent agreement with the experimental

| Phases  | Space group | Magnetism | Lattice parameters ($\text{Å}$) | k-point mesh | Refs. |
|---------|-------------|-----------|-------------------------------|--------------|------|
| Fe$_3$Sn | P6$_3$/mmc  | FM        | 5.457 4.362                   |              | Ref. 58 |
| Fe$_3$Sn | P6$_3$/mmc  | FM        | 5.461 4.347                   |              | Ref. 59 |
| Fe$_3$Sn | R-3 m       | FM        | 5.421 4.372                   |              | Ref. 60 |
| Fe$_3$Sn | R-3 m       | FM        | 5.440 4.352                   |              | Ref. 61 |
| Fe$_3$Sn | P6$_3$/mmc  | FM        | 5.464 4.352                   |              | Ref. 19 |
| Fe$_3$Sn | P6$_3$/mmc  | FM        | 5.475 4.307                   | 10 $\times$ 10 $\times$ 12 | This work |
| Fe$_3$Sn$_2$ | P6$_3$/mmc | FM        | 4.223 5.253                   |              | Ref. 62 |
| Fe$_3$Sn$_2$ | R-3 m    | FM        | 5.344 19.845                  |              | Ref. 63 |
| Fe$_3$Sn$_2$ | P6$_3$/mmc | FM        | 5.340 19.797                  |              | Ref. 19 |
| Fe$_3$Sn$_2$ | P6$_3$/mmc | FM        | 5.315 19.703                  |              | Ref. 3 |
| Fe$_3$Sn$_2$ | P6$_3$/mmc | FM        | 5.328 19.804                  | 10 $\times$ 10 $\times$ 3 | This work |
| Fe$_3$Sn$_2$ | P6$_3$/mmc | AFM       | 5.307 4.445                   |              | Ref. 58 |
| Fe$_3$Sn$_2$ | P6$_3$/mmc | AFM       | 5.297 4.481                   |              | Ref. 64 |
| Fe$_3$Sn$_2$ | P6$_3$/mmc | AFM       | 5.288 4.420                   |              | Ref. 65 |
| Fe$_3$Sn$_2$ | P6$_3$/mmc | AFM       | 5.300 4.450                   |              | Ref. 66 |
| Fe$_3$Sn$_2$ | P6$_3$/mmc | AFM       | 5.298 4.448                   |              | Ref. 19 |
| Fe$_3$Sn$_2$ | P6$_3$/mmc | AFM       | 5.297 4.449                   |              | Ref. 67 |
| Fe$_3$Sn$_2$ | I4/mcm     | AFM       | 5.299 4.449                   | 10 $\times$ 10 $\times$ 10 | This work |
| Fe$_3$Sn$_2$ | I4/mcm     | AFM       | 6.502 5.315                   |              | Ref. 68 |
| Fe$_3$Sn$_2$ | I4/mcm     | AFM       | 6.539 5.325                   |              | Ref. 69 |
| Fe$_3$Sn$_2$ | I4/mcm     | AFM       | 6.539 5.325                   |              | Ref. 70 |
| Fe$_3$Sn$_2$ | I4/mcm     | AFM       | 6.542 5.326                   |              | Ref. 64 |
| Fe$_3$Sn$_2$ | I4/mcm     | AFM       | 6.542 5.386                   |              | Ref. 65 |
| Fe$_3$Sn$_2$ | I4/mcm     | AFM       | 6.526 5.323                   |              | Ref. 19 |
| Fe$_3$Sn$_2$ | I4/mcm     | AFM       | 6.533 5.320                   |              | Ref. 71 |
| Fe$_3$Sn$_2$ | I4/mcm     | AFM       | 6.545 5.326                   |              | Ref. 72 |
| Fe$_3$Sn$_2$ | I4/mcm     | AFM       | 6.561 5.338                   | 8 $\times$ 8 $\times$ 10 | This work |
Fig. 1 Phonon dispersions of the pure elements and intermetallic phases in the Fe–Sn system. The black solid points represent the experimental data from ref. 37.
More interestingly, the magnetic contribution to the heat capacity presents at the magnetic phase transition of BCC-Fe. These results prove the accuracy of the current methods and justify the following calculations for intermetallics. Using the same strategy, we calculate heat capacities of Fe$_3$Sn$_3$, Fe$_5$Sn$_2$, Fe$_3$Sn, FeSn$_2$, and FeSn at finite temperatures, as shown in Fig. 2, with the magnetic heat capacity evaluated using Inden model. The heat capacity of Fe$_3$Sn shows a good consistency between our calculations and experiments at low temperature, which also confirms the accuracy of current theoretical results. We note that such good agreements are supported by considering the magnetic contributions in the magnetic system.

After getting the thermodynamical properties of intermetallics, we used CALPHAD method to evaluate the thermodynamic model parameters of the Fe–Sn system, and the phase diagram and thermodynamic properties are calculated by Thermo-Calc. Combining DFT and CALPHAD methods has already been successfully applied in different systems. Supplementary Table 1 lists the modeled thermodynamic parameters of the Fe–Sn system. The calculated Fe–Sn phase diagram is presented in Fig. 3 along with the experimental data. The comparison of the calculated temperatures and compositions of invariant reactions with experimental data as well as results from previous thermodynamic assessments are listed in Table 2.
Using the reactive crucible melting (RCM) approach, it is found that 3 metastable intermetallic compounds, i.e., Fe$_3$Sn, Fe$_5$Sn$_3$, and Fe$_2$Sn$_2$, can be stabilized between 873 K and 1173 K. Furthermore, we are convinced that the phase diagram reported in the literature is inaccurate in the temperature interval 1023–1038 K and Fe$_2$Sn can exist at 1023 K. Thus, the metastable phase Fe$_3$Sn is introduced by considering the current accurate experimental results. Obviously, good agreement between the optimized and introduced by considering the current accurate experimental data.

Our calculations demonstrate that Fe$_3$Sn exhibits the largest AHC and ANC values are promising candidates for transverse thermoelectric devices.

Symmetry plays a crucial role in determining the shape of the AHC and ANC tensors. AHC and ANC strongly depend on the Berry curvature according to:

$$\Delta \Omega (r) = \pm \text{det} (D(R) \cdot D(R) \cdot s^{-1} r),$$

where $\Omega (r)$ denotes the pseudovector Berry curvature, $D(R)$ the three-dimensional representation of a symmetry operation without the translation part and $s$ an arbitrary symmetry operation. That is, the symmetry operations of the magnetic point group will govern the shape of the tensors. Particularly, the ferromagnetic Fe$_3$Sn belongs to the magnetic space groups $Cmc'm'$ (BNS: 63.463), $Cm'c'm'$ (BNS: 63.464) and $P6_3/mmm'$ (BNS: 194.270) for the magnetic moments of Fe atoms pointing along the [100]-, [010]- and [001]-axis, respectively. Hence the presence of the 2$\sigma$, 2$\tau$, and 2$\rho$ rotation axes for each of magnetic space groups transform the Berry curvature according to:

For 2$\sigma$ with $M_{[100]}$

$$\Omega_\sigma (k_x, -k_y, -k_z) = -\Omega_\sigma (k_x, k_y, k_z)$$

$$\Omega_\sigma (k_x, -k_y, -k_z) = -\Omega_\sigma (k_x, k_y, k_z)$$

$$\Omega_\sigma (k_x, -k_y, -k_z) = -\Omega_\sigma (k_x, k_y, k_z)$$

For 2$\tau$ with $M_{[010]}$

$$\Omega_\tau (-k_x, k_y, -k_z) = -\Omega_\tau (k_x, k_y, k_z)$$

$$\Omega_\tau (-k_x, k_y, -k_z) = -\Omega_\tau (k_x, k_y, k_z)$$

$$\Omega_\tau (-k_x, k_y, -k_z) = -\Omega_\tau (k_x, k_y, k_z)$$

For 2$\rho$ with $M_{[001]}$

$$\Omega_\rho (-k_x, k_y, k_z) = \Omega_\rho (k_x, k_y, k_z)$$

$$\Omega_\rho (-k_x, k_y, k_z) = \Omega_\rho (k_x, k_y, k_z)$$

$$\Omega_\rho (-k_x, k_y, k_z) = \Omega_\rho (k_x, k_y, k_z)$$

The summation over the whole Brillouin zone forces $\sigma$, $\sigma$, and $\sigma$ for the magnetization direction along the [100]-axis to vanish, and

**Fig. 3** The optimized Fe–Sn phase diagram based on our thermodynamic modeling. The points correspond to the different experimental data. The red lines indicate the reaction temperatures between the different phases.
| Invariant reaction                                      | Reaction type | Composition at % Sn | Temperature (K) | Refs. |
|--------------------------------------------------------|---------------|---------------------|-----------------|-------|
| Liquid#1 → BCC_A2 + Liquid#2                          | Eutectic      | 0.312 0.083 0.811   | 1395.9          | Ref. 57 |
|                                                        |               |                     | 1381            | Ref. 45 |
|                                                        |               |                     | 1404            | Ref. 46 |
|                                                        |               |                     | 1403            | Ref. 47 |
|                                                        |               |                     | 1403            | Ref. 54 |
|                                                        |               |                     | 1407            | Ref. 35 |
|                                                        |               |                     | 1413            | Ref. 48 |
| BCC_A2 + Liquid → Fe₅Sn₃                               | Peritectic    | 0.297 0.095 0.796   | 1436            | This work |
|                                                        |               |                     | 1174.1          | Ref. 57 |
|                                                        |               |                     | 1166            | Ref. 48 |
|                                                        |               |                     | 1168            | Ref. 35 |
|                                                        |               |                     | 1183            | Ref. 50 |
|                                                        |               |                     | 1183            | Ref. 49 |
| BCC_A2 + Fe₅Sn₃ → Fe₅Sn₆                               | Peritectic    | 0.081 0.929 0.375   | 1111            | This work |
|                                                        |               |                     | 1174.1          | Ref. 57 |
|                                                        |               |                     | 1166            | Ref. 48 |
|                                                        |               |                     | 1168            | Ref. 35 |
|                                                        |               |                     | 1183            | Ref. 50 |
|                                                        |               |                     | 1183            | Ref. 49 |
| Fe₅Sn₃ + Liquid → Fe₅Sn₆                               | Peritectic    | 0.375 0.967 0.400   | 1074.8          | This work |
|                                                        |               |                     | 1072            | Ref. 57 |
|                                                        |               |                     | 1079            | Ref. 50 |
|                                                        |               |                     | 1079            | Ref. 49 |
|                                                        |               |                     | 1080            | Ref. 35 |
| Fe₅Sn₃ → Fe₅Sn + Fe₅Sn₆                               | Eutectoid     | 0.375 0.250 0.400   | 1062            | This work |
|                                                        |               |                     | 1024.7          | Ref. 57 |
|                                                        |               |                     | 1013            | Ref. 47 |
|                                                        |               |                     | 1034            | Ref. 35 |
|                                                        |               |                     | 1043            | Ref. 50 |
|                                                        |               |                     | 1043            | Ref. 49 |
| Fe₅Sn + Liquid → FeSn                                 | Eutectoid     | 0.400 0.980 0.500   | 874.9           | This work |
|                                                        |               |                     | 870             | Ref. 57 |
|                                                        |               |                     | 873             | Ref. 51 |
|                                                        |               |                     | 880             | Ref. 47 |
|                                                        |               |                     | 880             | Ref. 49 |
|                                                        |               |                     | 880             | Ref. 35 |
| Fe₅Sn + Liquid → FeSn                                 | Eutectoid     | 0.400 0.999 0.666   | 844             | This work |
|                                                        |               |                     | 775.4           | Ref. 57 |
|                                                        |               |                     | 769             | Ref. 48 |
|                                                        |               |                     | 769             | Ref. 45 |
|                                                        |               |                     | 769             | Ref. 47 |
|                                                        |               |                     | 786             | Ref. 49 |
|                                                        |               |                     | 786             | Ref. 50 |
|                                                        |               |                     | 786             | Ref. 35 |
| FeSn + Liquid → FeSn₂                                 | Peritectic    | 0.500 0.666 0.400   | 783             | This work |
|                                                        |               |                     | 784             | Ref. 57 |
|                                                        |               |                     | 769             | Ref. 48 |
|                                                        |               |                     | 769             | Ref. 45 |
|                                                        |               |                     | 769             | Ref. 47 |
|                                                        |               |                     | 786             | Ref. 49 |
|                                                        |               |                     | 786             | Ref. 50 |
|                                                        |               |                     | 786             | Ref. 35 |
| FeSn + Liquid → FeSn₂                                 | Eutectoid     | 0.999 0.666 0.333   | 504.9           | This work |
|                                                        |               |                     | 501             | Ref. 57 |
|                                                        |               |                     | 505             | Ref. 48 |
|                                                        |               |                     | 505             | Ref. 49 |
|                                                        |               |                     | 505             | Ref. 50 |
|                                                        |               |                     | 505             | Ref. 35 |
equivalently $\sigma_x$ and $\sigma_y$ and $\sigma_z$ for the magnetization along [010] and [001], respectively. However, there is no such condition for $\sigma_x$, $\sigma_y$, and $\sigma_z$ for magnetization direction along [100], [010], and [001] axes, respectively, and therefore they are allowed to have finite values.

AHC and ANC are proportional to the sum of the Berry curvature of the occupied bands, evaluated in the whole Brilouin zone (BZ), as defined in Eq (11). Since the Berry curvature depends on the energy difference between two adjacent bands, therefore it is expected that Weyl nodes as well as nodal lines, located close to the reference energy, contribute significantly to the total value, as shown in refs. 70,71 and confirmed for MnZn$^{62}$ and Mn$_3$PdN$^{72}$, respectively. Explicit band structure search reveals the presence of numerous Weyl nodes and nodal lines within the shaded energy range $[-0.118, -0.018]$ eV of Fig. 4a that are expected to contribute to the total AHC value. In order to identify the origin of the AHC contribution, we split the BZ into 216 cubes, within which the AHC is evaluated (see Fig. 4d). Since the major contribution originates from the diagonals, located within $k_z \in (-0.166, 0.000)$ (and $k_z \in (0.000, 0.166)$) as illustrated in Fig. 4d, it is fruitful to investigate the band gap within this $k_z$ range. Taking as an example the $k_z = -0.131$ plane, we plot the difference of the two involved bands as a black and white plot where the black areas correspond to small gap regions whereas white areas to large gap regions (see Fig. 4c). The shape of the gap plot is in complete agreement with the distribution of the AHC within the specified area, demonstrating that small gap regions similar to those within the square $k_x, k_y \in (0.333, 0.500)$ of Fig. 4c, contribute dominantly to the total AHC value.

Interesting topological transport properties can arise away from the charge-neutral point. One important observation is that the AHC curve of Fe$_3$Sn exhibits a sharp peak of $1308 \text{ S cm}^{-1}$ located at 60 meV below the Fermi level, as shown in Fig. 4. Therefore, an interesting question is whether tuning
the Fermi level to match the position of the peak is doable by means of doping. In order to investigate this possibility, we consider \((\text{Fe}_{1-x}\text{Mn}_x)_3\text{Sn}\) for various values of \(x\), with \(x \geq 0.15\), indicating the percentage of Mn doping to the system. By using virtual crystal approximation (VCA) calculations, we compute the AHC curve for different \(x\), as illustrated in Fig. 5a. It is noted that the position of the peak approaches the Fermi level while the Mn dopant concentration is increased and it hits the Fermi energy at approximately \(x = 0.15\) (black curve). The existence of the AHC peak and its location affects the calculated ANC. While the energy of the peak is lower than the Fermi energy (\(x < 0.15\)), the ANC is gradually decreased from \(-2.71 \text{ A m}^{-1} \text{ K}^{-1}\) for \(x = 0\) to \(-1.58 \text{ A m}^{-1} \text{ K}^{-1}\) for \(x = 0.15\). Once the energy of the peak gets larger than the Fermi energy (\(x > 0.15\)), ANC changes sign and jumps to \(3.63 \text{ A m}^{-1} \text{ K}^{-1}\) for \(x = 0.2\). Figure 5b shows the calculated ANC curves for various \(x\), demonstrating that Fe\(_3\)Sn offers an interesting playground of controlling the ANC by doping even with a sign change.

Since \((\text{Fe}_{0.85}\text{Mn}_{0.15})_3\text{Sn}\) exhibits the closest peak to the Fermi energy, its dynamical stability was checked by calculating its phonon dispersion (see Supplementary Fig. 3). To mimic the disorder structure, \((\text{Fe}_{0.85}\text{Mn}_{0.15})_3\text{Sn}\), a supercell, containing 80 atoms in special quasi-random structure (SQS)\(^{73}\), was generated by the mcsqs code of the ATAT package\(^{74}\). As illustrated in Supplementary Fig. 3, the absence of imaginary modes indicates \((\text{Fe}_{0.85}\text{Mn}_{0.15})_3\text{Sn}\) is dynamically stable.

Tuning the magnetization direction allows easier ANC modifications. In an attempt to tune the AHC and ANC of Fe\(_3\)Sn, we considered different magnetization directions i.e., along [100], [010], and [001] axes. Our results show no impact of the magnetization direction to the AHC and ANC values along [100] and [010] axis, where the values remain practically unchanged at 757 S cm\(^{-1}\) and \(-2.58 \text{ A m}^{-1} \text{ K}^{-1}\) due to the underlying hexagonal symmetry. On the other hand, a small change is noticed for direction along [001], where the AHC (ANC) is tuned to 676 S cm\(^{-1}\) (\(-2.06 \text{ A m}^{-1} \text{ K}^{-1}\)), see Fig. 5c and d (solid curves). Despite the minor changes in the AHC and ANC values at Fermi energy, a larger impact of the altering of the magnetization direction is observed away from the charge-neutral point for both magnetization directions and doping concentrations (solid and dashed curves, respectively). Specifically, the AHC peak of 1308 S cm\(^{-1}\) at 60 meV below the Fermi energy is moved closer to the Fermi energy, at 35 meV below the Fermi energy, and further reduces its maximum value to 886 S cm\(^{-1}\) when the magnetization direction is along the [001]-axis. The outcome of this change is more obvious in the ANC, where the zero value of the [001] direction is located closer to the Fermi energy, being useful for future applications. It is finally noted that for \(M||[001]\), doping results in ANC sign change (Fig. 5d, red curves).

Based on DFT calculations, the thermodynamical properties of the Fe–Sn system and the topological transport properties of Fe\(_3\)Sn are studied. Thermodynamic modeling of the Fe–Sn phase diagram has been re-established. The problems concerning
invariant reactions of intermetallics are remedied under our newly measured temperature ranges. First-principles phonon calculations with the QHA approach were performed to calculate the thermodynamic properties at finite temperatures. Thermodynamic properties, phonon dispersions of pure elements, and intermetallics were predicted to make up the shortage of experimental data. A set of self-consistent thermodynamic parameters are obtained by the CALPHAD approach. Further, we evaluated the AHC and ANC of Fe$_3$Sn with magnetization direction and doping being perturbations. The calculated AHC of 757 S cm$^{-1}$ is the largest among all reported members of the Fe-Sn family. It is noted that the nodal lines combined with the extended small gap areas constitute the main contribution to the total AHC and they can further be tuned by doping Mn at the Fe sites, allowing the manipulation of the AHC and ANC values and offering good candidate materials for promising transverse thermoelectric devices. In addition, promising high-throughput calculations\(^7^5,\)\(^7^6\) can be performed to search for more intriguing magnetic intermetallic compounds with singular topological transport properties, assisted by automated Wannier function construction\(^7^7\) for transport property calculations.

**METHODS**

**First-principles calculations**

Our calculations were performed using the generalized gradient approximation (GGA) for the exchange-correlation functional, in the parameterization of Perdew–Burke–Ernzerhof\(^7^8\) for the Vienna ab initio Simulation Package (VASP)\(^7^9,\)\(^8^0\). The energy cutoff is set at 600 eV and at least 5000 k-points in the first Brillouin zone with Γ-centered k-mesh were used for the hexagonal lattices (Fe$_3$Sn, FeSn, and Fe$_5$Sn$_3$), while for all the other structures, Monkhorst-Pack grids were used. The energy convergence criterion was set as 10$^{-6}$ eV, and 10$^{-5}$ eV Å$^{-1}$ was set as the tolerance of forces during the structure relaxation. The enthalpy of formation, $\Delta H (\text{Fe}_3\text{Sn}_y)$, for the Fe$_3$Sn$_y$ intermetallic compounds was obtained following

$$\Delta H (\text{Fe}_3\text{Sn}_y) = E_{\text{Fe}_3\text{Sn}_y} - \frac{x}{x+y} E_{\text{Fe}} - \frac{y}{x+y} E_{\text{Sn}},$$

where all the total energies for the equilibrium phases in their corresponding stable structures were obtained after structural relaxation.

For the phonon calculations, the frozen phonon approach was applied using the PHONOPY package.\(^8^0\) The temperature-dependent thermodynamical properties are calculated by using the quasi-harmonic approximation.\(^8^1\) The Gibbs free energy $G(T, P)$ at temperature $T$ and pressure $P$ can be obtained from the Helmholtz free energy $F(T, V)$ as follows:\(^8^2\)

$$G(T, P) = F(T, V) + PV = E_0(V) + F_{\text{el}}(V, T) + F_{\text{el}}(V, T) + F_{\text{mag}}(V, T),$$

where $E_0(V)$ is the total energy at zero Kelvin without the zero-point energy contribution, which were determined by fitting of the energies with respect to the volume data using the Birch–Murnaghan equation of state (EOS).\(^8^3\) $F_{\text{el}}$ corresponds to the lattice vibration contribution to the Helmholtz energy, which can be derived from the phonon density of states (PhDOS), $g(\omega, V)$, by using the following equation:\(^8^2\)

$$F_{\text{el}}(V, T) = k_B T \int_{0}^{\infty} 2 \sinh \frac{\omega}{2k_B T} g(\omega, V) d\omega,$$

where $k_B$ and $h$ are the Boltzmann constant and reduced Planck constant, respectively, and $\omega$ denotes the phonon frequency for a given wave vector $q$. The PhDOS $g(\omega, V)$ can be obtained by integrating the phonon dispersion in the Brillouin zone. The third term $F_{\text{mag}}$ represents the electronic contribution to the Helmholtz free energy, obtained by\(^8^4\):

$$F_{\text{el}}(V, T) = E_{\text{el}}(V, T) - T S_{\text{el}}(V, T)$$

where $E_{\text{el}}(V, T)$ and $S_{\text{el}}(V, T)$ indicate the electronic energy and electronic entropy, respectively. With the electronic DOS, both terms can be formulated as\(^8^4\):

$$E_{\text{el}}(V, T) = \int n(e) f_{\text{D}}(e) dE$$

$$S_{\text{el}}(V, T) = -k_B \int n(e) f_{\text{D}}(e) \ln f_{\text{D}}(e) dE,$$

where $n(e)$ is the electronic DOS, $f_{\text{D}}$ represents the Fermi-Dirac distribution function and $e_F$ is the Fermi energy.

Finally, based on the original Inden–Hillert–Jarl (IHQ) model\(^3^8,\)\(^3^9\) and further improved expression by Xiong,\(^8^5\) the magnetic Gibbs energy can be formulated as:

$$G_{\text{mag}} = RT (e_F^+ + 1) f(T),$$

where $\tau = T/T_c$, $T_c$ is the critical temperature (the Curie temperature $T_c$ for ferromagnetic materials or the Neel temperature $T_N$ for antiferromagnetic materials). $e_F^+$ is the effective magnetic moment per atom. And the relative parameters are summarized in Supplementary Table 2. Note that, we adopted the experimental critical temperatures and calculated magnetic moments.

In order to evaluate AHC, we projected the Bloch wave functions onto maximally localized wannier functions (MLWF) using Wanner90, following ref.\(^8^5\). A total number of 124 MLWFs, originating from the s, p and d orbitals of Fe atoms and the s and p orbitals of Sn atoms, are used. AHC is obtained by integrating the Berry curvature according to the formula:

$$\alpha_{\text{el}} = -\frac{c}{\pi} \int \frac{d\mathbf{k}}{2\pi} \sum_{\alpha\beta} f_{\alpha\beta}^{(k)} - \mu \Omega_{n,\alpha\beta}(k),$$

$$\Omega_{n,\alpha\beta}(k) = -2\Re \sum_{\alpha\beta} \frac{\langle k\mid \sum_{m} \left( n_{\alpha m}^* \mid k \right) \left( n_{\alpha m} \mid k \right) \rangle}{\left| \langle \beta\mid \sum_{m} \left( n_{\beta m}^* \mid k \right) \left( n_{\beta m} \mid k \right) \rangle \right|},$$

with $\mu$, $f$, $n$, $m$, $e_n(k)$, $e_m(k)$, and $\Omega_{n,\alpha\beta}(k)$ being the Fermi level, the Fermi-Dirac distribution function, the occupied Bloch band, the empty Bloch band, their corresponding energy eigenvalues and the Cartesian component of the velocity operator. The integration is performed on a $270 \times 270 \times 350$ mesh using WannierTools.\(^8^6\) ANC is evaluated using an in-house developed Python script, following the formula:

$$\alpha_{\text{ ANC}} = -\frac{1}{\pi} \int \frac{d\mathbf{q}}{2\pi} \frac{\partial f}{\partial \mathbf{q}} \sigma_{\text{ ANC}}(e) \epsilon - \mu \frac{\epsilon^2}{T},$$

where $T$, $e$, and $\epsilon$ are the temperature, the electronic charge and the energy point within the integration energy window, respectively. An energy grid of 1000 points within the window $[-0.5, 0.5]$ eV with respect to the Fermi level was chosen.

Mn doping at Fe sites is performed by using the virtual crystal approximation (VCA) as implemented in VASP.\(^8^7,\)\(^8^8\). In this approximation, virtual fictitious atoms that behave in between the parent atoms are inserted. VCA techniques have been used to describe prototypical doped $x$-FeMn systems\(^9^0\) and magnetic anisotropy energy of L1$_2$FePt and Fe$_{1-x}$Mn$_x$Pt\(^9^1\) and additionally topological transport properties of Co$_3$Ni$_2$Sn$_2$S$_2$\(^9^2\) and Fe$_3$Co alloys.\(^9^3\)

**CALPHAD modeling**

**Pure elements.** The Gibbs free energies for pure Fe and Sn were taken from the Scientific Group Thermodata Europe (SGTE) pure element database, which was described by:

$$G(T) = G(T) - H_{\text{mol}}(298.15K) = a + bT + cT^2 + dT^3 + eT^4 + fT^5 + gT^6 + hT^7 + i,$$

where $G(T)$ is the Gibbs free energy, $H_{\text{mol}}(298.15K)$ is the enthalpy of formation at 298.15 K, and $a, b, c, d, e, f, g, h, i$ are the material parameters.
where \( i \) represents the pure elements Fe or Sn, \( H_{\text{SER}}(298.15\text{K}) \) is the molar enthalpy of element \( i \) at 298.15 K in its standard element reference (SER) state, and \( a \) and \( b \) are known coefficients.

**Solution phases.** The solution phases, Liquid, BCC_A2, FCC_A1 and BCT_A5 phases are described using the substitutional solution model, with the corresponding molar Gibbs free energy formulated as:

\[
G_m^\phi = x_{\text{Fe}}G_{\text{Fe}}^\phi(T) + x_{\text{Sn}}G_{\text{Sn}}^\phi(T) + RT(x_{\text{Fe}}x_{\text{Fe}} + x_{\text{Sn}}x_{\text{Sn}}) + G^{\phi\phi} + G^{magn},
\]

(14)

where \( x_{\text{Fe}} \) and \( x_{\text{Sn}} \) are the mole fraction of Fe and Sn in the solution, respectively. Taken from SGTE94, \( G_{\text{Fe}}^\phi \) denotes the molar Gibbs free energy of pure Fe and Y in the structure \( \phi \) at the given temperature. \( G^{\phi\phi} \) denotes the excess Gibbs energy of mixing, which measures the deviation from the actual solution from the ideal solution behavior, modeled using a Redlich-Kister polynomial:

\[
G^{\phi\phi} = x_{\text{Fe}}x_{\text{Sn}} \sum_{j=0}^{n} \beta_j(x_{\text{Fe}} - x_{\text{Sn}})^j.
\]

(15)

The \( j \)th interaction parameter between Fe and Sn is described by \( \beta_j \), which is modeled in terms of a \( a+bT \).

**Stoichiometric intermetallic compounds.** FeSn_3, FeSn_2, FeSn, and FeSn_2 were considered as stoichiometric phases. The Gibbs free energies per mole atom of these phases were thus expressed as follows:

\[
G_m^{\text{FeSn}_3} = \frac{x}{x+y} G_{\text{Fe-SER}} + \frac{y}{x+y} G_{\text{Sn-SER}} + \Delta G_{\phi}^{\text{FeSn}_3}(T),
\]

(16)

where \( \Delta G_{\phi}^{\text{FeSn}_3}(T) \) is the Gibbs free energy of formation of the stoichiometric compound FeSn_3 which can be expressed as:

\[
\Delta G_{\phi}^{\text{FeSn}_3}(T) = A_3 + B_3 T.
\]

(17)

where the coefficients \( A_3 \) and \( B_3 \) are the parameters to be optimized. Since there is no experimental data of the thermodynamic properties for such intermetallic phases, the calculated enthalpies of formation for these phases from DFT calculations were treated as initial values of the coefficient \( A_3 \) in Eq. (17) in the present optimization.

**DATA AVAILABILITY**

The authors declare that the code supporting this study is available on GitHub under the link https://github.com/TMM-TUDA/Automatic-wannier-flow.

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COMPETING INTERESTS
The authors declare no competing interests.

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