Large anomalous Nernst effect and nodal plane in an iron-based kagome ferromagnet

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Anomalous Nernst effect (ANE), converting a heat flow to transverse electric voltage, originates from the Berry phase of electronic wave function near the Fermi energy $E_F$. Thus, the ANE provides a sensitive probe to detect a topological state that produces large Berry curvature. In addition, a magnet that exhibits a large ANE using low-cost and safe elements will be useful to develop a novel energy harvesting technology. Here, we report our observation of a high ANE exceeding 3 microvolts per kelvin above room temperature in the kagome ferromagnet Fe$_3$Sn with the Curie temperature of 760 kelvin. Our theoretical analysis clarifies that a “nodal plane” produces a flat hexagonal frame with strongly enhanced Berry curvature, resulting in the large ANE. Our discovery of the large ANE in Fe$_3$Sn opens the path for the previously unexplored functionality of flat degenerate electronic states and for developing flexible film thermopile and heat current sensors.

INTRODUCTION

Berry curvature, the fictitious magnetic field in momentum space, is the origin of various intrinsic transverse effects such as anomalous Hall effect (AHE), anomalous Nernst effect (ANE), and magneto-optical effects (1–15). Great efforts have been made to enhance Berry curvature not only by the way of the first-principles calculations using the modern high-throughput methods (16) but also by introducing external conditions to split the degeneracy of the bands, for example, the centrosymmetry breaking induced by strain, resulting in the substantial net Berry curvature (17). Recent progresses have revealed that the intrinsic ANE can be enhanced by the crossing of nondegenerate flat bands producing nontrivial topological structure, such as the Lifshitz transitions between type-I and type-II Weyl semimetals and nodal web, leading to the ever-recorded largest value of $6 \mu$V K$^{-1}$ at room temperature in Co$_2$MnGa and Fe$_3$X ($X = Ga$ and Al) (10, 11, 14, 15, 18). Following the concept behind these nodal points and lines produced by the band crossings (19), it would be highly interesting to consider even higher-dimensional crossings of the nondegenerate band producing a two-dimensional plane. Such a “nodal plane” would also have more advantages, by maximally contributing to the effective energy integral paths and by thus enhancing the Berry curvature and density of the states for spin up/down at the Fermi level in the first Brillouin zone (BZ) (10, 15). However, the formation of such a nontrivial nodal plane near the Fermi level has never been studied to date.

RESULTS

Here, we report the experimental observation of a room temperature high ANE and AHE in an iron-based binary ferromagnet based on the kagome lattice. Our comprehensive studies using both experiment and theory reveal the nodal plane formed in the kagome magnet, Fe$_3$Sn. This material has a hexagonal D0$_{19}$ type structure with the space group of $P6_3/mmc$, the same as the antiferromagnetic Weyl semimetals Mn$_3$X ($X = Sn$ or Ge), but differs from its two sibling compounds, tin-intercalated Fe-Sn binary compounds Fe$_3$Sn$_2$ and FeSn (Fig. 1A) (4–6, 20–25). In the ab plane, iron atoms form a kagome lattice, while the tin atoms occupy the center of each hexagon. Stacking two kagome layers with the half c axis separation and 60° rotation composes the primitive unit cell (Fig. 1A). This material has never been studied from the thermoelectric perspective, practically because of the difficulty of the synthesis (20, 26). Previous polycrystalline Fe$_3$Sn of hexagonal phase was synthesized by solid-state reaction method, and the product was very porous (20). In our study, we fabricated polycrystalline samples of Fe$_3$Sn D0$_{19}$ phase by arc melting method followed by a long period of annealing procedure (see Materials and Methods). Comprehensive transport measurements show a large Nernst coefficient, reaching up to $3 \mu$V K$^{-1}$ around room temperature, 10 times higher than that in pure iron. Moreover, our first-principles calculations confirm that the large transverse thermoelectric conductivity, $a_{ji}$, comes from the Berry curvature notably enhanced along a flat hexagonal frame formed by a nodal plane. Besides, this system is made of naturally abundant, low-cost element and has a high Curie temperature of 760 K and thus is useful for designing the transverse thermoelectric conversion device (Fig. 1B).
comparable to the maximum value reported for the Weyl ferromagnet Co₃Sn₄S₂ single crystal at 80 K and the same order of the magnitude as those reported for the topological ferromagnets Fe₃X and Co₃MnGa single crystals (10, 14, 15, 18, 27). To clarify the mechanism behind the observed large values, we estimate the transverse thermoelectric conductivity $\sigma_{ji}$ evaluated using the following equation, $\alpha_{ji} = \sigma_{ji} S_{ji} - \sigma_{ji} S_{ji}$, where $\sigma_{ji}$ is the longitudinal conductivity and Seebeck coefficient, respectively. As shown in Fig. 1D (blue line, right axis), $\alpha_{ji}$ linearly increases with $T$ at low temperatures, corresponding to $-\alpha_{ji}/T \sim 0.02$ A K⁻¹ m⁻¹, peaks at 200 K with the maximum value of $|\alpha_{ji}|_{\text{max}} \sim 2.5$ A K⁻¹ m⁻¹ and decreases to 2.2 A K⁻¹ m⁻¹ at 390 K. The values of $|\alpha_{ji}|/T$ and $|\alpha_{ji}|_{\text{max}}$ are comparable to the other topological magnets such as Co₃MnGa ($|\alpha_{ji}|_{\text{max}} \sim 4$ A K⁻¹ m⁻¹ (150 K), $|\alpha_{ji}|/T \sim 0.08$ A K⁻² m⁻¹ (10)), Fe₃Ga ($|\alpha_{ji}|_{\text{max}} \sim 5.2$ A K⁻¹ m⁻¹ (200 K), $|\alpha_{ji}|/T \sim 0.05$ A K⁻² m⁻¹ (15), and Co₃Sn₄S₂ ($|\alpha_{ji}|_{\text{max}} \sim 2.5$ A K⁻¹ m⁻¹ (150 K), $|\alpha_{ji}|/T \sim 0.03$ A K⁻² m⁻¹ (27)).

Next, we discuss the AHE. Figure 2A shows the field dependence of the Hall resistivity obtained at room temperature. In contrast with the Nernst coefficient, the Hall resistivity nearly saturates at the field around 0.1 T and still linearly increases with the magnetic field, likely due to the p-type character of the charge carrier at high temperatures. The Hall resistivity shows the monotonic increase with temperature and exceeds 3 microhm·cm above 300 K (Fig. 2B). The field dependence of the anomalous Hall conductivity (AHC) $-\sigma_{ji} = \rho_{ji}/\rho_{Lji}^2$ indicates a relatively large saturation value of about $-500$ S cm⁻¹ at 200 K (inset of Fig. 2A), again comparable to the reported $\sigma_{ji}$ in the cubic Fe₃X, indicating large net Berry curvature of the occupied bands. Moreover, the slope of $\sigma_{ji}$ changes systematically on cooling, which might arise from two possible sources: (i) the crossover from the p-type to n-type charge carriers that changes the sign of the ordinary Hall effect and (ii) the temperature variation of the anomalous Hall component that is directly related to the net magnetization $M$. As shown in Fig. 2C, the Hall conductivity $-\sigma_{ji}$ peaks at 200 K and gradually decreases on heating, reaching $-400$ S cm⁻¹ around 400 K. Figure 2D provides the temperature dependence of the transverse thermoelectric conductivity divided by temperature, $-\sigma_{ji}/T$. In the low-temperature range below $-40$ K, $-\sigma_{ji}/T$ is constant at $-0.02$ A K⁻² m⁻¹, followed by a logarithmic decrease with temperature nearly over a decade of $T$ up to 400 K, indicating the existence of nontrivial electronic structures close to the Fermi energy (12, 18).

Intrinsic anomalous Hall and Nernst effects both come from the Berry curvature in momentum space, but they have different dependence on the Fermi energy. Namely, AHE represents the integral of the Berry curvature over all the occupied bands, while the ANE measures the energy derivative of the AHC at the Fermi energy. We estimate the Fermi level by comparing the experimental results for AHC and ANE with the first-principles calculations based on density functional theory (DFT). The details of the calculation for the AHC and anomalous Nernst conductivity are shown in the
Supplementary Materials. The solid lines in Fig. 2 (C and D) show the best fits for the observed temperature dependence of AHC and $\alpha_{ji}/T$, which render the energy position for the polycrystalline Fe$_3$Sn, at $E_F = +88$ meV. Here, $E_F$ is the energy position for the theory formulated for a single crystalline Fe$_3$Sn. We note that this energy shift is consistent with the estimate made using the slight off-stoichiometry of the composition (Fe:Sn = 2.97:1.03) revealed by our chemical analysis (see Materials and Methods). The experimental AHC lies below the theoretical value in the whole temperature range as shown in Fig. 2C. Our analysis of $\sigma_{xx}$ dependence of $|\sigma_{xy}|$ indicates that this small deviation may originate from the extrinsic contribution (see text S5 and fig. S7), and thus, the majority part is based on the intrinsic mechanism.

To investigate the origin of AHE and ANE, we calculate the AHC and anomalous thermoelectric conductivity based on DFT. For a polycrystalline sample of Fe$_3$Sn, we estimate the AHC and transverse thermoelectric conductivity as an average of $\sigma_{ji}$ and $\alpha_{ji}$ for each magnetization direction along principal crystal axes, [21T0], [01T0], and [0001] (see Materials and Methods and text S4). Figure 3 (A and B) shows the chemical potential dependence of the AHC and anomalous thermoelectric conductivity for Fe$_3$Sn. AHC has a sharp peak around $E \sim +50$ meV as shown in Fig. 3A. This peak is related to the odd-functional form of $-\alpha_{ji}^{\text{avg}}/T$ showing two sharp peaks with opposite signs at $E \sim +30$ and $+80$ meV, as the thermoelectric conductivity $\alpha_{ji}$ can be evaluated as the energy derivative of AHC. The structure becomes sharper on cooling to the $T = 0$ K limit (fig. S4, B and C). To clarify the origin of the peak, we focus on the analysis of the in-plane Berry curvature $\mathbf{\Omega}_x$ at $E \sim +50$ meV as this peak of AHC comes from the in-plane components ($\sigma_{xz}$ and $\sigma_{zx}$) (fig. S4A). As we will discuss below, we find strongly enhanced Berry curvature on a particular (nodal) plane by monitoring $\mathbf{\Omega}_x$ over all BZ. Figure 3C shows the sum of the Berry curvature over the occupied states on the $k_z = 0.13$ plane. The strongly enhanced Berry curvature appears on the lines connecting each $U^*$ point at $E \sim +50$ meV (Fig. 3C, left). On the other hand, the Berry curvature vanishes at $E \sim +90$ meV (Fig. 3C, right). Note that each $U^*$ point is located slightly inside the BZ boundary.

It is known that the Berry curvature takes a large value at band crossing points and enhances AHE and ANE. In the absence of the spin orbit coupling (SOC), the band degeneracy occurs accidentally when the two crossing bands have the same spin. If the degeneracy is protected by some symmetry of crystal and magnetic structure, then the band crossing points form a nodal line. On the other hand, when the degeneracy is made from up- and down-spin bands, the crossing points make a surface, which we shall call nodal plane. This is because there is no hybridization between up- and down-spin.
bands, so that there is only one constraint that defines the degeneracy. Figure 3D shows the nodal plane satisfying the following condition: 

$$\Delta \varepsilon(k) \equiv \varepsilon^\alpha(k) - \varepsilon^\beta(k) = 0,$$

where $\Delta \varepsilon(k)$, $\varepsilon^\alpha(k)$, and $k$ are the energy difference between two bands, eigenvalue for up- and down-spin bands, and wave vector, respectively. The color bar corresponds to the eigenvalue for the nodal plane. We see that the nodal plane has a constant eigenvalue ($E \sim +50$ meV) on the $U^*_1 - U^*_2$ and $U^*_4 - U^*_5 - U^*_6$ line, along which the Berry curvature is found to be strongly enhanced (Fig. 3C, left). This means that the nodal plane on the lines connecting each $U^*$ point has a flat energy dispersion.

This can be confirmed in the band structure on $U^*_1 - U^*_2$ line (Fig. 3E). The energy bands with a very small energy dispersion appear on the characteristic symmetry line connecting the $U^*$ points. All the analyses clarify that the partially flat bands with the nodal plane lead to the significant enhancement in AHC. This is fully consistent with recent works on the nearly flat dispersion region inducing the enhanced Berry curvature and ANE (10, 15, 28).

In general, nodal planes become nodal lines or Weyl points after introducing SOC. For the case of Fe$_3$Sn, however, the gap fully opens at any $k$ point at the $k_z = 0.13$ plane, and no Weyl points appear (Fig. S9, C to E). Instead, we found partially flat bands with a
small energy gap as shown in fig. S9E, which is the origin of the large ANE.

Similar to the nodal lines, nodal planes are also ubiquitous in electronic band structures without SOC. Nodal planes generally can become lines at a constant energy cut, while nodal lines become points (fig. S9, A and B). Thus, these constant energy lines in nodal planes become partially flat bands along this line with a small energy gap after introducing SOC. In addition to this general feature of nodal planes, the band structure that we focused on appears at the stationary point of the nodal plane, which may enhance the density of states and thus ANE (28).

This characteristic band structure is not unique for Fe3Sn and might be realized in other kagome lattice materials. In general, it is well known that M points in kagome lattice make van Hove singularity (a derivative of energy dispersion is 0). Therefore, energy dispersion on lines connecting M points could also be flat comparing with other k path. The partially flat bands in Fe3Sn might come from the symmetry of kagome lattice because U*1−U*2 line is the same direction as the k path connecting the M points.

DISCUSSION

Many criteria are important to evaluate the performance of a transverse thermoelectric material. First of all, the high Nernst coefficient is inevitable for efficiently converting longitudinal heat current to transverse voltage drop. Second, such a high-ANE material must have a magnetic transition temperature much higher than room temperature, as most of waste heat is available at temperatures between room temperature and 200°C (29). Third, the materials should be made of the economically low-cost and safe materials to produce flexible films for two-dimensional coverage of a heat source.

Figure 4A shows the summary of the temperature dependence of the anomalous Nernst coefficient for each magnetic compound ever reported. Here, we can see that the most recently found topological magnets have much better performance such as the maximum ~3, 4, 6, and 8 μV K−1 in Co3Sn2S2 at 80 K and Fe3Al, Fe3Ga, and Co3MnGa at 400 K, respectively. Even in the antiferromagnets Mn3Sn and Mn3Ge, the corresponding ANE values, 0.6 and 1.35 μV K−1 (8, 21), are large enough, comparable with the values reported for conventional strong ferromagnets such as Co/Ni alloy, Mn3Ga, etc. For application, the binary magnets are better suited than ternary as the fabrication process becomes much simpler. In this sense, the most promising ANE compounds are the recently reported Fe3X alloys, featuring high-ANE performance and economically low-cost and safe materials for fabrication (17). However, tin is much cheaper than gallium, which provides an alternative selection for implementing transverse thermoelectric devices.

Figure 4B shows the full logarithmic plot of the anomalous Nernst coefficient versus magnetization for various kinds of magnets. Conventionally for ordinary ferromagnets, the ANE is proportional to its magnetization M and is confined in the yellow shaded region in Fig. 4B (1, 8, 30–32). For topological magnets, however, the ANE is much more enhanced than the empirical linear relation with M, and we note that the ANE in Fe3Sn (red closed star) just lies among these values reported for topological magnets (blue shaded region).

We have found a high intrinsic ANE at room temperature in the iron-based kagome ferromagnet Fe3Sn. Our comprehensive study using both experiment and theory finds that both ANE and AHE observed in experiment come from the nodal plane producing Berry curvature enhanced along a hexagonal frame near the Fermi energy. As almost all the applications are based on a polycrystalline sample, our discovery of the high-performance thermoelectricity in the polycrystalline form of Fe3Sn is crucial and would open various possible venues for future developments of flexible film thermopile and heat current sensors. Further study is necessary, especially to realize large ANE at zero magnetic field by enhancing the coercivity using the shape anisotropy in a thin-film form (33) or by introduction of pinning centers.

MATERIALS AND METHODS

Sample preparation and characterizations of the polycrystalline Fe3Sn

Polycrystalline Fe3Sn ingots were made by arc melting the mixtures of iron chops (4 N) and tin grains (5 N) under an argon atmosphere for oxygen extinction. After being sealed into an ampoule and annealed for 7 days at 805°C, the polycrystalline ingot was quenched into ice water (26). The shining sample was cut into several parts by a spark-erosion machine for varieties of characterizations. The x-ray powder diffraction data were taken by “x-ray fluorescence reduction mode” of a Rigaku D/teX250 diffractometer (SmartLab, Rigaku Corp.), which can reduce the background from the fluorescent effect. The Rietveld refinement analysis indicates that the main phase is Fe3Sn and the inclusion of impurity phase Fe5Sn3 (~7 weight%). This is consistent with the estimate from the magnetization anomaly due to the magnetic ordering. The atomic ratio of iron to tin was confirmed to be 2.97:1.03, very close to stoichiometric 3:1, by inductively coupled plasma (ICP) method. This slight off-stoichiometry leads to 0.18 electron doping per formula unit by counting the number of d and p electrons for the outermost shell. Electron doping of 0.18 corresponds to £9 ~ +90 meV, which is consistent with the estimate of the Fermi energy shift made using αxy/T and σxy. Magnetization measurements were carried out on a commercial superconducting quantum interference device magnetometer [Magnetic Property Measurement System (MPMS), Quantum Design]. The specific heat measurement was performed using a commercial system [Physical Property Measurement System (PPMS), Quantum Design] under zero magnetic field (see fig. S1).

Electric and thermolectric measurements of the polycrystalline Fe3Sn sample

The high field Hall resistivity plotted in Fig. 2A was measured by a standard six-probe method using a commercial system (PPMS, Quantum Design). Electrical contacts with gold wires were made by epoxy bonding. The sample (~1 mm in length by ~0.3 mm in width by 0.1 mm in thickness) was fully fixed by varnish to prevent any movement under high magnetic fields. All other electric and thermal transport properties were measured by a thermal transport option on a commercial system (PPMS, Quantum Design) with a rectangular bar-shaped sample (~6 mm in length by ~1.3 mm in width by ~0.5 mm in thickness). One of the edges of the sample was clamped at the cold hearth, and a resistive heater is attached at the other edge. Two Cernox thermometers are also attached at the middle of the sample to measure temperatures of two different positions ΔT = Thot − Tcold. Using the distance x between the two thermometers, the thermal gradient can be calculated as ∆T/x. Here, AT typically set to be 1.5 to 2.0% of the sample temperature. The Hall signal can be recorded simultaneously, while thermoelectric
is being measured. The Seebeck coefficient $S_{ij}$ and Nernst signal $S_{ij}$ were then estimated as $S_{ij} = E_i/T + E_j/T$, where $E_i$ and $E_j$ are the longitudinal and transverse electric fields.

**DFT calculation**

The electronic structures of Fe$_3$Sn were calculated using the OpenMX code, where the exchange-correlation functional within the generalized-gradient approximation and norm-conserving pseudo-potentials were used (22). The SOC is included using total angular momentum–dependent pseudo-potentials (34, 35). The wave functions were expanded by a linear combination of multiple pseudo-atomic orbitals (36). A set of pseudo-atomic orbital basis was specified as Fe6.0-s3p3d3f1, Sn7.0-s3p2d2f1, where the number after each element stands for the radial cutoff in the unit of Bohr and the integer after s, p, and d indicates the radial multiplicity of each angular momentum component. The cutoff energy for a charge density of 300 rydbergs, p, and d indicates the radial multiplicity of each angular momentum component. The tesseract for a charge density of 300 rydbergs, p, and d indicates the radial multiplicity of each angular momentum component. The tesseract for a charge density of 300 rydbergs, p, and d indicates the radial multiplicity of each angular momentum component. The tesseract for a charge density of 300 rydbergs, p, and d indicates the radial multiplicity of each angular momentum component.

From the Bloch states obtained in the DFT calculation described above, a Wannier basis set was constructed using the Wannier90 code (37). The basis was composed of (d), character orbitals localized at each Fe, and (s, p), character ones at Sn site, which are 76 orbitals (38). The basis was composed of (d), character orbitals localized at each Fe, and (s, p), character ones at Sn site, which are 76 orbitals (38). The basis was composed of (d), character orbitals localized at each Fe, and (s, p), character ones at Sn site, which are 76 orbitals (38). A set of pseudo-atomic orbital basis was specified as Fe6.0-s3p3d3f1, Sn7.0-s3p2d2f1, where the number after each element stands for the radial cutoff in the unit of Bohr and the integer after s, p, and d indicates the radial multiplicity of each angular momentum component. The cutoff energy for a charge density of 300 rydbergs, p, and d indicates the radial multiplicity of each angular momentum component. The cutoff energy for a charge density of 300 rydbergs, p, and d indicates the radial multiplicity of each angular momentum component. The cutoff energy for a charge density of 300 rydbergs, p, and d indicates the radial multiplicity of each angular momentum component.

For all of these Wannier Hamiltonians, the intrinsic contributions of the AHC $\sigma_{ij}$ and the anomalous transverse thermoelectric conductivity $\alpha_{ij}$ were calculated through the Berry curvature formula as

$$\sigma_{ij}(T, \mu) = - \frac{e}{h} \int \frac{dk}{(2\pi)^3} \Omega_{\alpha\gamma}(k) f_n(k)$$

$$\alpha_{ij}(T, \mu) = - \frac{1}{e} \int d\epsilon \left( \frac{\partial f}{\partial \epsilon} \right) \sigma_{ij}(\epsilon, \mu) \frac{\epsilon - \mu}{T}$$

where $\epsilon$, $h$, $f$, $\mu$, $\epsilon_{ij\gamma}$, and $\Omega_{\alpha\gamma}$ are the elementary charge with a negative sign, the reduced Planck constant, the band energy, the Fermi–Dirac distribution function with the band index $n$ and the wave vector $k$, the chemical potential, Levi-Civita symbol, and the $\gamma$ component of the Berry curvature, respectively. We calculate an average for $\sigma_{ij}$ and $\alpha_{ij}$ to compare the theory to the experimental results for polycrystalline samples as $\sigma_{ij}^{xy} = \frac{1}{3}(\sigma_{xy} + \sigma_{yx} + \sigma_{yx})$ and $\alpha_{ij}^{xy} = \frac{1}{3}(\alpha_{xy} + \alpha_{yx} + \alpha_{yx})$ (see also text S4).

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