



This is the accepted manuscript made available via CHORUS. The article has been published as:

Robust Nernst magnetothermoelectricity in the topological spin semimetal $\text{FeCrRh}_2\text{X}_4$ ($\text{X} = \text{Si}, \text{Ge}$)

Amit Chanda, Jadupati Nag, Noah Schulz, Derick DeTelle, Aftab Alam, K. G. Suresh, Manh-Huong Phan, and Hariharan Srikanth

Phys. Rev. B **111**, 094416 — Published 17 March 2025

DOI: [10.1103/PhysRevB.111.094416](https://doi.org/10.1103/PhysRevB.111.094416)

Robust Nernst magneto-thermoelectricity in topological spin semi-metal FeCrRhX (X=Si, Ge)

Amit Chanda^{1,2*}, Jadupati Nag^{3,4}, Noah Schulz¹, Derick DeTellem¹, Aftab Alam³, K. G.

Suresh³, Manh-Huong Phan¹ and Hariharan Srikanth^{1*}

¹*Department of Physics, University of South Florida, Tampa FL 33620, USA*

²*Department of Energy Conversion and Storage, Technical University of Denmark,
2800 Kgs. Lyngby, Denmark*

³*Magnetic Materials Laboratory, Department of Physics, Indian Institute of
Technology Bombay, Mumbai - 400076, India*

⁴*Materials Research Institute and Department of Material Science & Engineering,
Pennsylvania State University, University Park, PA 16802, USA*

**Corresponding authors: amitch@dtu.dk, sharihar@usf.edu*

Abstract

We report on the anomalous transverse magneto-thermoelectric properties of the topological spin semi-metallic equiatomic quaternary Heusler alloys (EQHAs) FeCrRhSi and FeCrRhGe over a wide temperature range between 130 K and 295 K. Magnetic measurements reveal ferromagnetic behavior above room temperature with high Curie temperatures ($T_C > 350$ K) in these samples, where energetically competing ferromagnetic (FM) and antiferromagnetic (AFM) phases coexist at low temperatures, giving rise to field-induced metamagnetic transitions. The electrical resistivity of both samples exhibits semi-metallic characteristics throughout the measured temperature range, with low temperature resistivity upturns driven by Kondo-like scattering. Throughout this range, the longitudinal Seebeck coefficient is negative in sign for both FeCrRhSi and FeCrRhGe, indicating that electrons are the majority carriers for

thermally driven charge transport. Notably, both samples exhibit giant values of the anomalous Nernst coefficient (S_{zy}^{ANE}) at room temperature, with S_{zy}^{ANE} values of $(0.272 \pm 0.02) \mu\text{V} \cdot \text{K}^{-1}$ for FeCrRhSi and $(0.295 \pm 0.02) \mu\text{V} \cdot \text{K}^{-1}$ for FeCrRhGe. These values are nearly three times larger than those reported for other EQHAs in the literature. An in-depth analysis reveals that intrinsic Berry curvature drives such large anomalous Nernst effects (ANEs) in our EQHAs. We show that in these samples, the contribution of the transverse thermoelectric conduction to the observed large ANE dominates over that of anomalous Hall effect acting on the charge carrier flow driven by the longitudinal Seebeck effect. The ratio of the anomalous off-diagonal transverse thermoelectric conductivity (α_{zy}^A), to the anomalous Hall conductivity (σ_{zy}^{AHE}), $|\alpha_{zy}^A/\sigma_{zy}^{AHE}|$ lies between $0.2 \left(\frac{k_B}{e}\right)$ and $\left(\frac{k_B}{e}\right)$ at room temperature, further highlighting the intrinsic Berry curvature as the underlying physical origin of the observed large ANEs.

Keywords: Quaternary Heusler alloys, topological spin semi-metals, anomalous Nernst effect, Berry curvature, anomalous Hall effect.

1. Introduction

The global quest for sustainable energy resources and novel energy harvesting technologies has led to the emergence of spin caloritronics, [1] a field that combines spin transport and magneto-thermoelectricity wherein, the anomalous Nernst effect (ANE) [2] has become an efficient technique for exploiting the spin degree of freedom of an electron in addition to its charge degree of freedom to harvest clean electrical energy from renewable thermal energy sources. When a temperature gradient is applied to a magnetic conducting or semiconducting material with a spontaneous magnetization, an electric field is generated along a direction orthogonal to both the temperature gradient and the material's magnetization. This phenomenon is known as ANE where the thermally induced transverse electric field is proportional to the cross product of the thermal gradient and spontaneous magnetization of the material [3]. ANE-based thermoelectric (TE) devices bring several additional advantages as compared to conventional charge Seebeck effect-based TE devices, such as higher efficiency, simplified electrical connections for ANE-based thermopiles, scalability of the output voltage with the device dimensions enabling mass-production *etc.* [4,5] The physical mechanisms responsible for ANE can be categorized as either extrinsic or intrinsic in origin. While large Berry curvature at the Fermi level (E_F) mediated by the topologically non-trivial band structure is recognized as the intrinsic origin of ANE, [6–8] side-jump mechanism, [9–11] and asymmetric skew scattering of charge carriers [6,12] are some of the effects responsible for the extrinsic origin of ANE. Intrinsic Berry curvature-driven colossal ANE at room temperature has been observed in a wide range of magnetic topological materials, including the semi-metallic full Heusler alloy Co_2MnGa , [7,13] ferromagnetic Weyl semimetal $\text{Co}_3\text{Sn}_2\text{S}_2$, [8,14] Kagome magnets Fe_3Sn and Fe_3Sn_2 , [15,16] the cubic ferromagnets Fe_3Ga and Fe_3Al , [17] cubic magnetic alloy Fe_3Pt , [18] the chiral antiferromagnetic Mn_3Sn , [19], and the semi-metallic Heusler ferromagnet CoFeVSb , [20] *etc.* Extrinsic skew scattering-driven large ANE

has also been observed in ferromagnetic mixed-valent manganites such as $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ [21] and $\text{La}_{1-x}\text{Na}_x\text{MnO}_3$ [22], *etc.*, hole-doped ferromagnetic cobaltites like $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ [23–25], spin gapless semiconductors like $\text{Co}_{1-x}\text{Fe}_{1+x}\text{CrGa}$, [12] Cr-based equiatomic quaternary Heusler alloys (EQHAs) such as CrRuMnGe and CrRuCoGe , [26] the half metallic ferromagnet Fe_3O_4 [27]. It is important to note that intrinsic Berry curvature-driven ANE results in a higher value of the anomalous Nernst coefficient (ANC) compared to extrinsically driven ANE [4,28]. Furthermore, intrinsically driven ANE has been reported to scale beyond magnetization in topological materials with no net magnetization but a large net Berry curvature around the Fermi level. [7] Therefore, from the viewpoint of thermal energy harvesting, more specifically, for constructing ANE based thermopiles, [4,5] room temperature topological magnets with colossal ANC are preferred.

In this context, magnetic topological Heusler alloys hold great promise due to their high Curie temperatures (T_C), large spin polarization, and topologically non-trivial band structures. These properties have generated growing interest in these materials for potential spintronic and spin caloritronic applications [29,30]. Amongst the vast family of magnetic Heusler alloys, the full Heusler alloys such as Co_2MnGa , [7,13] Co_2MnAl [31] and Co_2MnSi , [32] $\text{Co}_2\text{MnAl}_{1-x}\text{Si}_x$, [33] $\text{Co}_2\text{Fe}_{0.4}\text{Mn}_{0.6}\text{Si}$ [34], Co_2TiSn [35], Ni_2MnGa , [36] Fe_2CoX ($X = \text{Al}$ and Ga), Fe_2NiX ($X = \text{Al}$ and Ga), [37] *etc.*, exhibit giant and promising ANC at room temperature. EQHAs with high spin polarization and high T_C are also known to exhibit versatile magneto-electronic properties *e.g.*, half-metallicity, [38,39] spin gapless semiconducting, [40–42] spin-valve, [43] spin semi-metallic and Weyl semi-metallic behavior, [44,45] topological spin semi-metallicity with fully compensated ferrimagnetism, [46] bipolar magnetic semiconducting, [47] as well as remarkable thermoelectric performances [12,20,38]. Despite their multifunctionality, studies on ANE in EQHAs are relatively scarce in contrast to full Heusler alloys. Our group has

reported large values of the ANE in several EQHAs, such as CrRuXGe ($X = \text{Co}$ and Mn), [26] spin gap-less semiconductors like $\text{Co}_{1-x}\text{Fe}_{1+x}\text{CrGa}$ [12,48], and the semi-metallic ferromagnet CoFeVSb [20]. Interestingly, while most of the aforementioned alloys exhibit positive ANE, CrRuMnGe exhibits negative ANE [26]. In majority of these EQHAs, the ANE is governed by the extrinsic skew scattering mechanism, with the exception of CoFeVSb where the intrinsic Berry curvature drives a significantly large ANE [20]. Since colossal values of ANC at room temperature are expected to be observed in topological magnets that facilitate large Berry curvatures at or near the E_F , [4,28] EQHAs with topologically nontrivial band structures appear to be promising candidates for efficient thermal to electrical energy conversion based on ANE.

Given the aforementioned promising properties of EQHAs, we have chosen to study two candidate materials FeCrRhSi and FeCrRhGe from this family, which are established to be spin-metals in our previous reports. [44,45] Spin semi-metals are recently discovered interesting family of magnetic materials with immense potential for room temperature spintronic applications. In these materials, one of the spin bands is semi-metallic due to the partial overlap of valence and conduction bands, which can occur through direct or indirect overlap, resulting in a large density of states (DOS) at E_F , whereas the other spin band is gapped, exhibiting insulating or semiconducting behavior. [44–46] In the spin semi-metallic band near the E_F , direct overlap between conduction and valence bands often leads to doubly degenerate, linear band crossings. [44] The schematic illustration of the spin resolved DOS for spin semi-metals is shown in **Fig. 1(a)** indicating overlapped spin-up and gapped spin-down bands at E_F . Note that the spin resolved DOS for spin semi-metals is unique and fundamentally different from that of half metallic ferromagnets and spin gapless semiconductors [12,48]. For comparison, the spin resolved DOS for half metallic ferromagnets and spin gapless semiconductors is presented in **Fig. 1(b)**, indicating the partially filled valence band at E_F for

the spin-up channel and gapped spin-down band for the half-metallic ferromagnets, whereas gapless spin-up bands and gapped spin-down bands at E_F for the spin gapless semiconductors. Band structure calculations have demonstrated that the semi-metallic spin-up band exhibits type-II Weyl semi-metallic behavior in FeCrRhGe. [44] Due to these unique topologically non-trivial band structures [44,45], both FeCrRhSi and FeCrRhGe exhibit sizeable anomalous Hall conductivity arising from their intrinsic Berry curvature. This finding was further confirmed by the *ab-initio* calculations and Hall data analysis, highlighting their potential for spintronic and spin caloritronic applications. This motivated us to take the next step and investigate their transverse thermo-spin effects. Here, we report the first experimental observation of ANE in FeCrRhSi and FeCrRhGe. We have obtained the large ANC values for both FeCrRhSi and FeCrRhGe at room temperature, which are nearly three times higher than those reported for other EQHAs in the literature. Our in-depth analysis indicates that the large ANE in both systems is governed by the intrinsic Berry curvature. The contribution of the transverse thermoelectric conduction to the observed anomalous Nernst thermoelectricity surpasses the contribution of the anomalous Hall effect acting on the charge carrier flow driven by the longitudinal Seebeck effect.

2. Experimental Details

Polycrystalline FeCrRhSi and FeCrRhGe alloys were prepared by arc melting technique as reported in previous studies [44,45]. The crystal structures of these alloys at room temperature were characterized by X-ray diffraction (XRD) pattern by employing a Xpert Pro diffractometer with Cu-K α radiation ($\lambda = 1.54184$ Å). Magnetic measurements of these alloys were conducted using a vibrating sample magnetometer (VSM) incorporated to a physical property measurement system (PPMS) from Quantum Design, USA. The longitudinal resistivity and Hall resistivity of these alloys were measured using the standard four-probe

technique by utilizing the DC resistivity option of the PPMS. The Seebeck coefficient and the total thermal conductivity of these alloys were simultaneously measured by employing the Thermal Transport Option (TTO) of the PPMS. Samples with dimensions $5 \times 4 \times 1.17 \text{ mm}^3$ (for FeCrRhSi) and $5 \times 4 \times 0.97 \text{ mm}^3$ (for FeCrRhGe) were used for the electrical and thermoelectric transport measurements. Anomalous Nernst measurements were performed as a function of temperature and magnetic field using a custom-built spin caloritronic set up integrated with the PPMS based on a universal sample puck for the PPMS. The samples were sandwiched between two copper plates in such a manner that a temperature gradient is applied along the thickness of the samples, and the direction of the temperature gradient is orthogonal to the direction of the external DC magnetic field provided by the PPMS. It should be noted that the bottom copper plate (hot) was thermally isolated from the base of the PPMS puck by a 4 mm thick Teflon block, whereas the top copper plate (cold) was thermally anchored to the base of the PPMS puck through a pair of molybdenum screws. The anomalous Nernst voltage generated in the samples while sweeping the external DC magnetic field and in presence of the applied temperature gradient was recorded by a Keithley 2182A nanovoltmeter. A more detailed description of the spin caloritronic set up is reported in our previous studies [12,20,26,48,49].

3. Results and Discussion

3.1. Structural Properties

The room temperature XRD patterns for both FeCrRhSi and FeCrRhGe samples used in this study are already reported in the literature by our group [44,45]. These materials belong to the class of EQHAs ($XX'YZ$) and adopt a cubic crystal structure with space group $F\bar{4}3m$ associated with the structural prototype of LiMgPdSn. In general, an EQHA $XX'YZ$ can stabilize in three energetically different non-degenerate structural configurations, namely, type I, type II and

type III, with different occupancies of Wyckoff positions $4a$ (0, 0, 0), $4b$ (0.5, 0.5, 0.5), $4c$ (0.25, 0.25, 0.25), and $4d$ (0.75, 0.75, 0.75), keeping the Z-atom fixed at the $4a$ site. [39] These three structural configurations can be visualized as, type I: X at the $4d$ site, X' at the $4c$ site, and Y at the $4b$ site; type II: X at the $4b$ site, X' at the $4c$ site, and Y at the $4d$ site; and type III: X at the $4c$ site, X' at the $4b$ site, and Y at the $4d$ site. Apart from this, there are possibilities of B2 or A2 type disorder as well, where two pairs or all 4-atomic sites prefer to remain in a disordered fashion. Both FeCrRhSi and FeCrRhGe are shown to crystallize in $L2_1$ structure with $Y = \text{Cr}$ at $4b$, $Z = \text{Si (Ge)}$ at $4a$, and 50% antisite disorder between the tetrahedral site atoms Fe and Rh (X and X') [44,45]. From the Rietveld analysis, the refined values of lattice parameters of FeCrRhSi and FeCrRhGe were found to be 5.90 Å and 5.79 Å, respectively [44,45]. As the XRD patterns for these samples have been reported in the literature over a broad range of the scattering angle ($20^\circ \leq 2\theta \leq 120^\circ$) [44,45], we only display the room temperature XRD patterns for FeCrRhSi and FeCrRhGe in **Figs. 1(c)** and **(d)**, respectively within a narrower range of scattering angles ($41^\circ \leq 2\theta \leq 47^\circ$) to feature the most intense (220) peak, located at $2\theta \approx 44.47^\circ$ for FeCrRhSi and 43.18° for FeCrRhGe. The average crystalline grain size (D) was estimated for both samples using the Scherrer equation, [50] $D = \frac{K\lambda}{W \cos \theta}$; where, λ is the wavelength of X-ray, θ is the diffraction angle, W is the full width at half maximum of the diffraction peak and K is the Scherrer constant. Considering $\lambda = 1.5418 \text{ Å}$, $K = 0.89$, [50] we obtained $D = (11.4 \pm 1.0)$ and $(19.2 \pm 2.0) \text{ nm}$ for FeCrRhSi and FeCrRhGe, respectively. **Figures 1(e)** and **(f)** display the schematic representations of the crystal structures of FeCrRhSi and FeCrRhGe, respectively considering type I structure [51,52], where Cr occupies the $4b$ site, Si (Ge) occupies the $4a$ site, and 50% antisite disorder exists between the tetrahedral site atoms Fe and Rh ($4c$ and $4d$) [44,45].

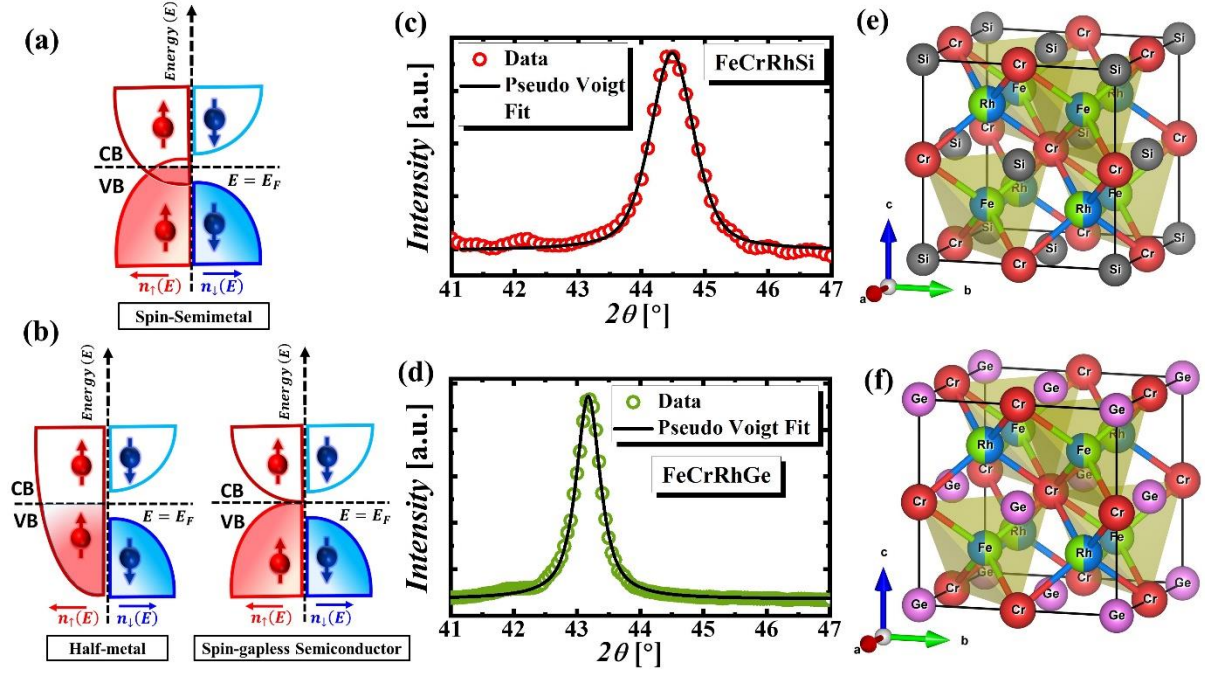


Figure 1 Schematic illustrations of the electronic density of states $n(E)$ for (a) spin semi-metals and (b) half metals and spin gapless semiconductors. Room temperature XRD patterns for (c) FeCrRhSi and (d) FeCrRhGe. Schematic representations of the crystal structures (top view) of (e) FeCrRhSi and (f) FeCrRhGe.

3.2. Magnetic Properties

Figures 2(a) and (b) depict the temperature dependence of magnetization, $M(T)$ of FeCrRhSi and FeCrRhGe, respectively measured in presence of an external magnetic field of $\mu_0 H = 0.05$ T under the zero-field cooled warming (ZFCW) and field cooled warming (FCW) protocols in the temperature (T) range: $10 \text{ K} \leq T \leq 390 \text{ K}$. Clearly, the magnetic transition temperatures (T_C) of both samples are above 350 K, indicating their above-room temperature ferromagnetic nature [44,45]. It seems that FeCrRhGe has a higher T_C than FeCrRhSi as both the ZFCW and FCW $M(T)$ curves for FeCrRhGe continue to increase with temperature up to 390 K, while those for FeCrRhSi start dropping abruptly above 350 K. To have a clear idea about the ferromagnetic transition in these alloys, the FCW $M(T)$ for FeCrRhGe was

measured at $T > 400$ K. The inset of **Fig. 2(b)** shows the FCW $M(T)$ of FeCrRhGe measured in the presence of an external magnetic field of $\mu_0 H = 0.05$ T over the temperature range of 450 K to 700 K indicating $T_C = 550$ K for this sample [44]. Therefore, the T_C of FeCrRhSi is estimated to lie in the temperature range: $400 \text{ K} \leq T_C \leq 550 \text{ K}$. Since we do not have the high temperature $M(T)$ data for FeCrRhSi, we fitted the ZFCW $M(T)$ for FeCrRhSi with an extended form of the Bloch's law [53–56]: $M(T) = M(T_0) \left[1 - \left(\frac{T-T_0}{T_C-T_0} \right)^\beta \right]$; where $M(T_0)$ is the value of M at $T = T_0$, β is the Bloch's exponent, and $T_0 = 300$ K is the lowest temperature for our selected fitting region ($T_0 \leq T \leq T_C$) where the ferromagnetic interactions play a dominating role. From the extended Bloch's law fitting of the FCW $M(T)$ data for FeCrRhSi (shown in the inset of **Fig. 2(a)**), we obtained $T_C = (458 \pm 1) \text{ K}$, which agrees with our prediction about the T_C of this sample. Furthermore, we obtained $\beta = 4.22 \pm 0.30$, which corresponds to 3D isotropic exchange interactions with integer spin quantum number ($\beta = 9/2$). [53,55] In order to verify the compatibility of the aforementioned Bloch's model to our EQHAs, we have performed the same fitting of the FCW $M(T)$ data for FeCrRhGe (shown in the inset of **Fig. 2(b)**), and obtained $T_C = (565 \pm 1) \text{ K}$ and $\beta = 4.21 \pm 0.10$. Thus, the value of T_C obtained for the FeCrRhGe from the Bloch's law fit matches closely with the experimental value of T_C . Furthermore, the obtained value of β is close to $\beta = 9/2$ indicating 3D isotropic exchange interactions in case of FeCrRhGe alloy as well. [53,55] It is also noteworthy that the FCW $M(T)$ curve for both these samples bifurcate slightly from the ZFCW $M(T)$ below 300 K. Moreover, ZFCW and FCW $M(T)$ for both these samples decrease slowly with decreasing temperature up to $T = 50$ K below which both ZFCW and FCW $M(T)$ show a slope change. Such behavior of the ZFCW and FCW $M(T)$ curves has also been observed in the semi metallic EQHA CoFeVSb, [20,43] and can be associated with the coexistence of energetically competing ferromagnetic (FM) and antiferromagnetic (AFM) phases driven by

the antisite disorder present in the FeCrRhSi and FeCrRhGe samples [44,45]. Such antisite disorder mediated concomitance of energetically competing magnetic phases is responsible for the bifurcation in ZFCW and FCW $M(T)$, [20,43] which is more pronounced below 50 K for both samples. Previous *ab-initio* calculations suggest that both long-range super-exchange AFM interactions between the Cr atoms and the antisite disorder between Fe and Rh atoms contribute to the resulting AFM interactions [44,45]. It is also worth noting that the $M(T)$ values are consistently higher in FeCrRhGe (**Fig. 2(b)**) than in FeCrRhSi (**Fig. 2(a)**) at all temperatures and under the same applied magnetic field strength.

Main panels of **Figs. 2(c)** and **(d)** demonstrate the magnetic field dependence of magnetization, $M(H)$ for the FeCrRhSi and FeCrRhGe samples, respectively at $T = 300$ and 3 K. The saturation magnetization (M_S) at $T = 3$ K for the FeCrRhSi and FeCrRhGe are $M_S = 2 \mu_B/\text{f. u.}$ and $2.9 \mu_B/\text{f. u.}$, respectively. It is well-known that the magnetic moment of Heusler alloys can be predicted by estimating the total number of valence electrons (N_V). Since $N_V = 27$ for both FeCrRhSi and FeCrRhGe, according to the Slater-Pauling rule, [57] the theoretically estimated value of $M_S = (N_V - 24) \mu_B/\text{f. u.} = 3 \mu_B/\text{f. u.}$ for both samples. Therefore, it is evident that the discrepancy between the theoretically estimated and experimentally observed M_S value is greater for FeCrRhSi than for FeCrRhGe. This indirectly hints towards a higher degree of inherent antisite disorder in FeCrRhSi, leading to stronger competition between the FM and AFM phases, and consequently, enhanced magnetic frustration at low temperatures.

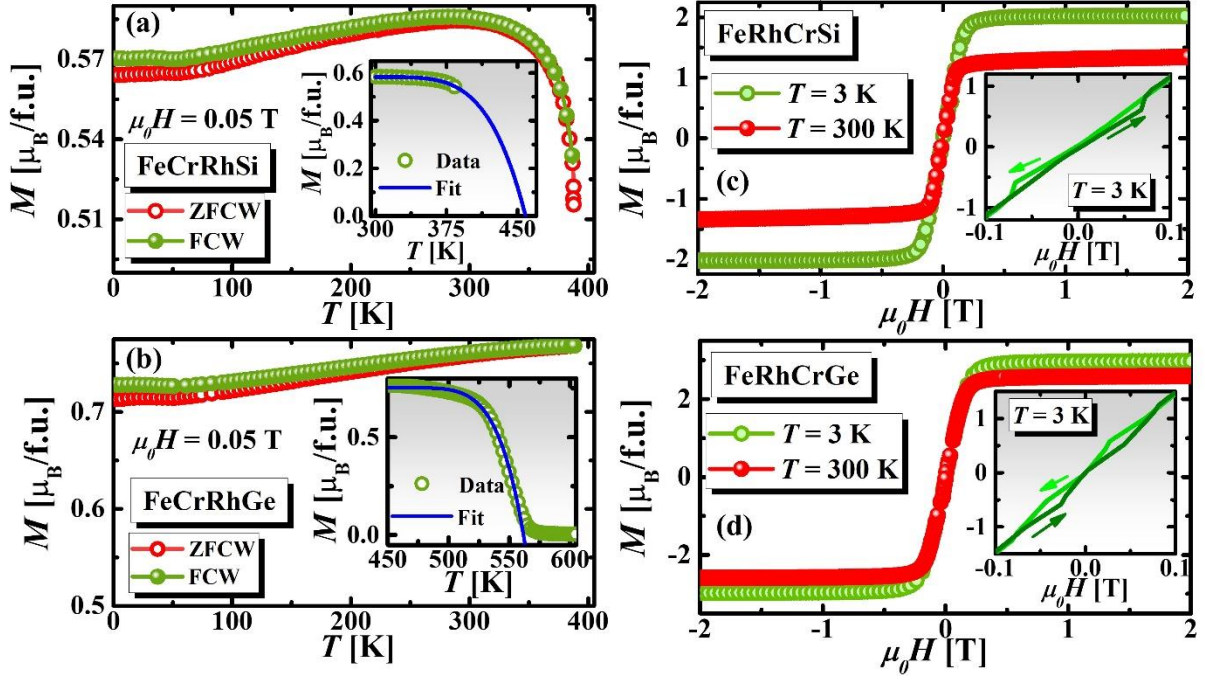


Figure 2. Temperature dependence of magnetization, $M(T)$ of (a) FeCrRhSi and (b) FeCrRhGe, measured in the presence of an external magnetic field of $\mu_0 H = 0.05$ T under the zero-field cooled warming (ZFCW), and field cooled warming (FCW) protocols in the temperature range: $10 \text{ K} \leq T \leq 390 \text{ K}$. Insets show the Bloch's law fitting of the FCW $M(T)$ data for FeCrRhSi and FeCrRhGe, respectively. (c, d) Magnetic field dependence of magnetization, $M(H)$ for FeCrRhSi and FeCrRhGe at 300 and 3 K. Insets show their low field $M(H)$ behavior at $T = 3 \text{ K}$.

To gain a deeper insight into this, we have taken a closer look at the low field behavior of the $M(H)$ loops ($-0.1 \text{ T} \leq \mu_0 H \leq +0.1 \text{ T}$) for both samples at $T = 3 \text{ K}$. As shown in the insets of **Fig. 2(c)** and (d), the $M(H)$ curves exhibit a remarkable low field feature, with a pronounced magnetization switching close to the zero field for both the forward and reverse branches of the $M(H)$ loop leading to the appearance of a wasp-waist shaped hysteresis loop. Such a behavior has been observed in compensated ferrimagnetic $\text{Fe}_{100-x}\text{Gd}_x$ alloy films [58], magnetic shape memory alloy $\text{Ni}_{50}\text{Mn}_{34}\text{In}_{16}$, [59] magnetically inhomogeneous Mn_3GaC [60],

EQHA CrRuMnGe [26] *etc.* Such wasp-waist shaped $M(H)$ loops in the low field regime for our FeCrRhSi and FeCrRhGe samples at low temperatures are likely to arise from the magnetic field-induced metamagnetic transition from the AFM to the FM state. Notably, the critical field for this metamagnetic transition (H_{CM}) is higher in the FeCrRhSi sample ($H_{CM} \approx \pm 0.068$ T) compared to the FeCrRhGe sample ($H_{CM} \approx \pm 0.042$ T). This suggests a stronger AFM phase fraction in FeCrRhSi, which explains the greater suppression of magnetization in FeCrRhSi than in FeCrRhGe at low temperatures.

3.3. Electrical and Thermoelectric Transport Properties

Main panels of **Figs. 3(a)** and **(b)** show the temperature dependence of longitudinal electrical resistivity, $\rho_{xx}(T)$ for the FeCrRhSi and FeCrRhGe samples, respectively in the temperature range: $2 \text{ K} \leq T \leq 300 \text{ K}$ for FeCrRhSi and $3 \text{ K} \leq T \leq 300 \text{ K}$ for FeCrRhGe. It is evident that $\rho_{xx}(T)$ exhibits negative temperature coefficient ($\frac{\partial \rho_{xx}}{\partial T} < 0$) throughout the measured temperature range for both samples indicating their semiconducting or semi-metallic behavior [44,45]. However, the absence of exponential temperature dependence of ρ_{xx} rules out the possibility of the gapped semiconducting nature for both the samples. Furthermore, the order of magnitude and the temperature dependence of ρ_{xx} resemble those of semi-metallic systems [44,45]. Interestingly, $\rho_{xx}(T)$ curves for these samples exhibits an evident slope-change at low temperatures (see insets of **Fig. 3(a)** and **(b)**), followed by a resistivity upturn with further lowering of temperature. Such resistivity upturn at low temperatures has been observed in various Heusler alloy systems *e.g.*, the full Heusler alloys $\text{Fe}_2\text{VAl}_{1-x}\text{Si}_x$, [61] $\text{Fe}_{2-x}\text{Cr}_x\text{Val}$, [62] *etc.*, which usually arises as a result of scattering of conduction electrons by the localized magnetic impurity potential. This phenomenon is recognized as the Kondo-like scattering [63] which is experimentally manifested as a minimum in $\rho_{xx}(T)$ with a $-\ln T$ dependence just below the minimum. Notably, while the resistivity upturn in FeCrRhSi appears

below 20 K that for FeCrRhGe appears below 10 K indicating stronger Kondo-like scattering in FeCrRhSi in comparison to FeCrRhGe.

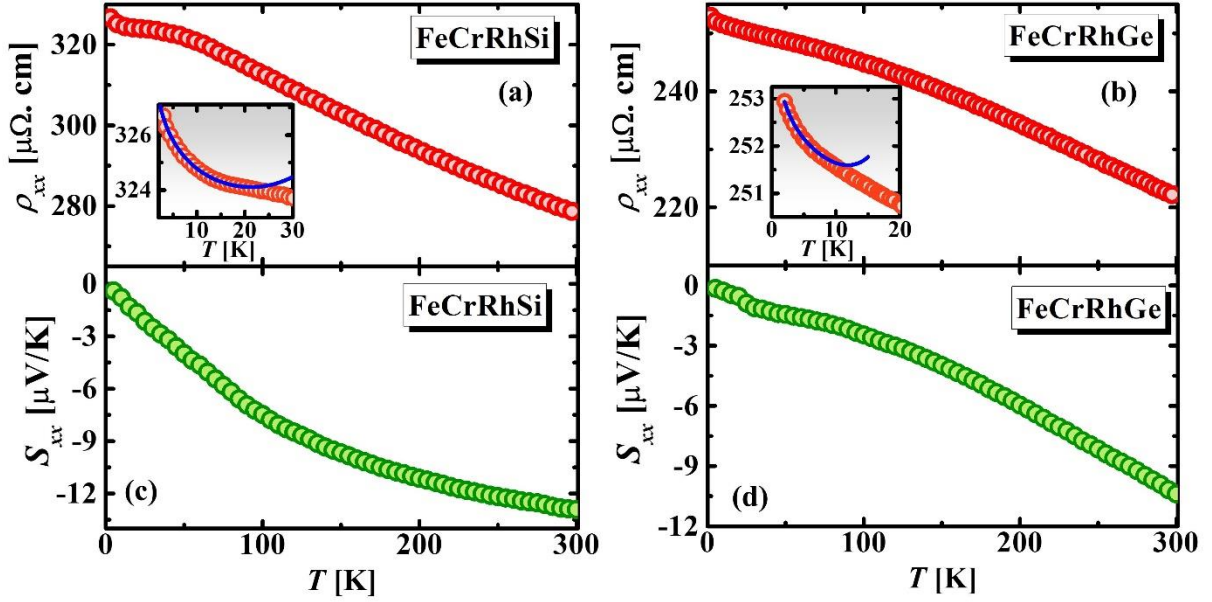


Figure 3. Temperature dependence of longitudinal resistivity, $\rho_{xx}(T)$ for (a) FeCrRhSi and (b) FeCrRhGe fitted with the expression: $\rho_{xx}(T) = \rho_0 - \rho_K \ln T + \rho_{e-ph} T^5 + \rho_{e-e} T^2$ in the low temperature region (insets). Temperature dependence of longitudinal Seebeck coefficient S_{xx} for (c) FeCrRhSi and (d) FeCrRhGe.

To gain a deeper insight into the electrical transport properties of these FeCrRhSi and FeCrRhGe EQHAs at low temperatures, we have analyzed the $\rho_{xx}(T)$ data, considering different bulk scattering mechanisms. The temperature dependence of ρ_{xx} of a disordered conducting magnetic system can be expressed as a linear combination of several contributions, $\rho_{xx}(T) = \rho_0 + \rho_{xx}^K(T) + \rho_{xx}^{in-el}(T)$ [64,65]. Here, ρ_0 accounts for the temperature independent residual resistivity, $\rho_{xx}^K(T)$ represents the Kondo-like scattering (*i.e.*, $\rho_{xx}^K(T) = -\rho_K \ln T$), [61] and ρ_{xx}^{in-el} indicates the combined contribution of various inelastic scattering channels, *e.g.*, electron-electron (ρ_{xx}^{e-e}), electron-phonon (ρ_{xx}^{e-ph}), and electron-magnon (ρ_{xx}^{e-m}) scatterings, *etc.* [65] The electron-electron scattering originates from the inelastic

collision between the s -electrons and d -electrons and typically shows a T^2 dependence (i.e., $\rho_{xx}^{e-e}(T) = \rho_{e-e}T^2$). [66,67] On the other hand, the contribution of electron-magnon or, single-magnon scattering also shows a T^2 dependence (i.e., $\rho_{xx}^{e-m}(T) = \rho_{e-m}T^2$) [65]. However, similar to the half metallic ferromagnets, since there is a gap in the DOS for the minority spin sub-band, only one spin channel (majority spin-up channel) contributes to the electrical transport in case of spin-semimetals, and hence single-magnon scattering can be ruled out [68]. Therefore, we have neglected the contribution of single-magnon scattering in our analysis. Finally, the electron-phonon scattering contribution towards ρ_{xx} is typically governed by the Bloch-Grüneisen model, [65,69,70] $\rho_{xx}^{e-ph}(T) = \rho_{BG} \left(\frac{T}{\theta_D}\right)^m \int_0^{\frac{\theta_D}{T}} \frac{e^{x^m} dx}{(e^x - 1)^2}$. Here, ρ_{BG} is a constant pre-factor, θ_D is the Debye temperature, and m is a constant exponent which is usually ≈ 5 for a conducting material. For the sake of simplicity, we have considered, $\rho_{xx}^{e-ph}(T) = \rho_{e-ph}T^5$. Considering the aforesaid scattering mechanisms, the $\rho_{xx}(T)$ data was found to fit well with the expression, [71] $\rho_{xx}(T) = \rho_0 - \rho_K \ln T + \rho_{e-ph}T^5 + \rho_{e-e}T^2$ in the temperature regions: $2 \text{ K} \leq T \leq 20 \text{ K}$ for FeCrRhSi and $3 \text{ K} \leq T \leq 10 \text{ K}$ for FeCrRhGe, respectively. The fitting parameters for both the samples are presented in **Table 1**. It is evident that the contribution of the Kondo-like scattering in ρ_{xx} is higher in the FeCrRhSi alloy in comparison to the FeCrRhGe alloy, which agrees with our experimental observation of (1) stronger upturn in $\rho_{xx}(T)$ in case of FeCrRhSi in contrast to that in FeCrRhGe and (2) appearance of the upturn in $\rho_{xx}(T)$ at a higher temperature in FeCrRhSi in comparison to FeCrRhGe. Nonetheless, the enhanced Kondo-like scattering in FeCrRhSi may arise from the higher degree of antisite disorder in FeCrRhSi as compared to FeCrRhGe. [72] It is also noteworthy that the contribution of electron-phonon scattering towards electrical transport is higher in FeCrRhGe compared to FeCrRhSi.

Next, we discuss the longitudinal thermoelectric transport properties of the FeCrRhSi and FeCrRhGe samples. **Figures 3(c)** and (d) display the temperature dependence of longitudinal charge Seebeck coefficient, $S_{xx}(T)$ for the FeCrRhSi and FeCrRhGe samples, respectively. Evidently, the sign of $S_{xx}(T)$ is negative throughout the measured temperature range for both FeCrRhSi and FeCrRhGe samples indicating that electrons are the majority carriers for the thermally driven charge transport in these samples. Notably, the absolute values of $S_{xx}(T)$ are slightly higher in FeCrRhSi than in FeCrRhGe at all temperatures, which is in agreement with a previous report [38]. Although the absolute value of S_{xx} decreases gradually with decreasing the temperature for both these samples, the behavior of the $S_{xx}(T)$ curve for FeCrRhSi is strikingly different from that of FeCrRhGe. While the slope of the $S_{xx}(T)$ curve increases gradually with decreasing temperature for FeCrRhSi, FeCrRhGe exhibits a reverse trend. Nevertheless, $S_{xx}(T)$ curves for both samples display a change of slope around 100 K, which is more pronounced in the FeCrRhSi alloy. In this context, it is also noteworthy that the observed negative sign of $S_{xx}(T)$ in both these alloys contradicts the positive sign of ordinary Hall coefficients found in these alloys [44,45]. This apparent inconsistency between the Seebeck coefficient and Hall resistivity has also been noticed in other Heusler alloys *e.g.*, Co₂MnGa [7], Fe-Based Heusler Compounds such as Fe₂NiAl [37]. These alloys also exhibit positive Hall coefficient but negative sign of Seebeck coefficient. We found that not only these but also the simplest metal like Lithium with negative charge carriers (electrons) shows positive Seebeck coefficient [73]. Using first principle calculations, it was shown that the sign of the Seebeck coefficient is determined by the energy dependence of the electron lifetime (which is generically proportional to the inverse of the electronic density of states) rather than the electron-phonon interactions [73]. Therefore, in order to explain the anomalous sign of the Seebeck coefficient in our FeCrRhSi and FeCrRhGe samples, detailed *ab initio* calculations within the framework of Boltzmann's transport theory need to be performed.

Parameters	FeCrRhSi	FeCrRhGe
ρ_0 ($\mu\Omega \cdot \text{cm}$)	(328 ± 0.07)	(253 ± 0.02)
ρ_K ($\mu\Omega \cdot \text{cm}$)	(1.65 ± 0.05)	(0.85 ± 0.2)
ρ_{e-ph} ($\mu\Omega \cdot \text{cm} \cdot \text{K}^{-5}$)	$(1.48 \pm 0.5) \times 10^{-8}$	$(6.8 \pm 0.7) \times 10^{-7}$
ρ_{e-e} ($\mu\Omega \cdot \text{cm} \cdot \text{K}^{-2}$)	$(1.45 \pm 0.06) \times 10^{-3}$	$(2.09 \pm 1) \times 10^{-3}$
v_S ($\text{m} \cdot \text{s}^{-1}$)	3050	4596
λ_{ph} (nm)	11.4	19.2
δ	188	152
A (s^3)	$(0.85 \pm 0.1) \times 10^{-43}$	$(4.05 \pm 0.1) \times 10^{-43}$
B ($\text{s} \cdot \text{K}^{-1}$)	$(1.12 \pm 0.05) \times 10^{-22}$	$(0.6 \pm 0.2) \times 10^{-22}$
C (s)	$(2.78 \pm 0.1) \times 10^{-15}$	$(2.52 \pm 0.5) \times 10^{-15}$
θ_D (K)	(370 ± 10)	(498 ± 10)

Table 1. List of parameters obtained by fitting the $\rho_{xx}(T)$ and $\kappa_L(T)$ data for the FeCrRhSi and FeCrRhGe alloys.

3.4. Thermal conductivity and thermoelectric figure of merit

Next, we shed some light on the longitudinal thermal transport properties of these samples. The left vertical axes of **Figs. 4(a)** and **(b)** represent the temperature dependence of total longitudinal thermal conductivity, $\kappa_{Tot}(T)$ in the temperature range: $5 \text{ K} \leq T \leq 300 \text{ K}$ for the FeCrRhSi and FeCrRhGe samples, respectively. The right vertical axes, on the other hand, display the thermoelectric figure of merit, $ZT = \frac{S_{xx}^2}{\kappa_{Tot}\rho_{xx}} T$ [74] for the same two samples. Note that ZT at 300 K is higher in FeCrRhSi ($ZT \approx 0.005$) in comparison to that of FeCrRhGe ($ZT \approx 0.003$), which is in agreement with earlier reports on ZT for these two samples. [38] In general, $\kappa_{Tot}(T)$ can be expressed as, $\kappa_{Tot}(T) = \kappa_{el}(T) + \kappa_L(T)$, where, $\kappa_{el}(T)$ and $\kappa_L(T)$

are the electronic and lattice contribution to the total thermal conductivity respectively. Since T_C of FeCrRhSi and FeCrRhGe is well above 400 K, we have neglected the magnon contribution towards κ_{Tot} for both the samples, as the magnons contribute only $\approx 5 - 10 \%$ of κ_{Tot} around T_C in majority of the magnetic conductors/semiconductors [75]. The left vertical axes of **Figs. 4(c)** and (d) display $\kappa_{el}(T)$ for FeCrRhSi and FeCrRhGe, respectively, estimated using the Wiedemann-Franz law: $\kappa_{el}(T) = \frac{L_0 T}{\rho_{xx}}$, where, $L_0 = \frac{\pi^2 k_B^2}{3e^2} = 2.44 \times 10^{-8} \text{ W. } \Omega. \text{ K}^{-1}$ is the Lorenz number for free electrons (degenerate limit) [76], and $\rho_{xx}(T)$ are the measured values of resistivity. It is important to note that L_0 for semiconductors with high value of S_{xx} deviates from the degenerate limit and becomes temperature dependent. In order to account for such deviations, a modified temperature dependent L_0 was proposed, which can be expressed as, [77] $L_0(T) = 1.5 + \exp[-|S_{xx}(T)|/116] \text{ W. } \Omega. \text{ K}^{-1}$. Therefore, we have estimated $\kappa_{el}(T)$ for both the samples using the expression: $\kappa_{el}(T) = \frac{[1.5 + \exp[-|S_{xx}(T)|/116]]T}{\rho_{xx}}$. The right vertical axes of **Figs. 4(c)** and (d) depict $\kappa_L(T)$ for the FeCrRhSi and FeCrRhGe samples, respectively estimated using the expression: $\kappa_L(T) = \kappa_{Tot}(T) - \kappa_{el}(T)$. Notably, $\kappa_L(T)$ for FeCrRhSi exhibits a sharp maximum at/around $\approx 100 \text{ K}$ whereas a comparatively broad shoulder at $\approx 125 \text{ K}$ for FeCrRhGe.

To analyze the contributions of different phonon scattering processes towards the lattice thermal conductivity, we fitted $\kappa_L(T)$ for FeCrRhSi and FeCrRhGe with the Debye-Callaway model, [78,79]

$$\kappa_L(T) = \frac{k_B}{2\pi^2 v_S} \left(\frac{2\pi k_B T}{h} \right)^3 \int_0^{(\theta_D/T)} \frac{\tau_L t^4 e^t}{(e^t - 1)^2} dt \quad (1)$$

Here, v_S is the average phonon velocity, $t = \frac{\hbar\omega}{k_B T}$, ω is the phonon frequency, and τ_L is the total phonon relaxation-time. Here, the reciprocal of τ_L can be written as a linear combination of

different phonon scattering processes as, $\tau_L^{-1} = \tau_B^{-1} + \tau_D^{-1} + \tau_U^{-1} + \tau_{e-ph}^{-1}$; where, τ_B^{-1} , τ_D^{-1} , τ_U^{-1} and τ_{e-ph}^{-1} represent the reciprocal of the relaxation times related to the phonon-boundary scattering, phonon scattering process by point defects, the Umklapp, and the electron-phonon scattering process, respectively [80]. For diffusive boundary scattering, $\tau_B^{-1} = \frac{v_s}{\delta \cdot \lambda_{ph}}$, where λ_{ph} is the mean free path of phonons and δ is the correction factor associated with the phonon scattering processes at the grain boundaries. For a polycrystalline sample, $\lambda_{ph} \approx$ the average grain size, or the particle size. For scattering of phonons by point defects, $\tau_D^{-1} = A\omega^4$, where the parameter A is a constant pre-factor that depends on the mass-fluctuation scattering and phonon speed [81]. The relaxation time for the three-phonon Umklapp process in Callaway's model is expressed as, [82] $\tau_U^{-1} = B'\omega^2 T^3 e^{-(\theta_D/T)}$. However, after comparing the thermal conductivities of different crystals, Slack *et al.*, [83,84] proposed the modified expression for the relaxation time for the Umklapp scattering process as [82,83], $\tau_U^{-1} = B\omega^2 T e^{-(\theta_D/3T)}$, where B is a constant pre-factor. Relaxation time associated with the electron-phonon scattering process takes the form, $\tau_{e-ph}^{-1} = C\omega^2$ [85]. We have considered the reported values of $v_s = 3050 \text{ m. s}^{-1}$ and 4596 m. s^{-1} for FeCrRhSi and FeCrRhGe, respectively [38], which are also close to the average v_s reported for other EQHAs, *e.g.*, CoFeVSb, [20] CrRuCoGe and CrRuMnGe, [26] *etc.* Our XRD analysis revealed that the average grain size, $D = (11.4 \pm 1.0) \text{ nm}$ and $(19.2 \pm 2.0) \text{ nm}$ for FeCrRhSi and FeCrRhGe, respectively. Therefore, we have considered $\lambda_{ph} = 11.4 \text{ nm}$ and 19.2 nm for the two alloy systems, and the best fits were obtained for $\delta = 188$ (152) for FeCrRhSi (FeCrRhGe). The values of the fitting parameters for FeCrRhSi and FeCrRhGe are shown in **Table 1**.

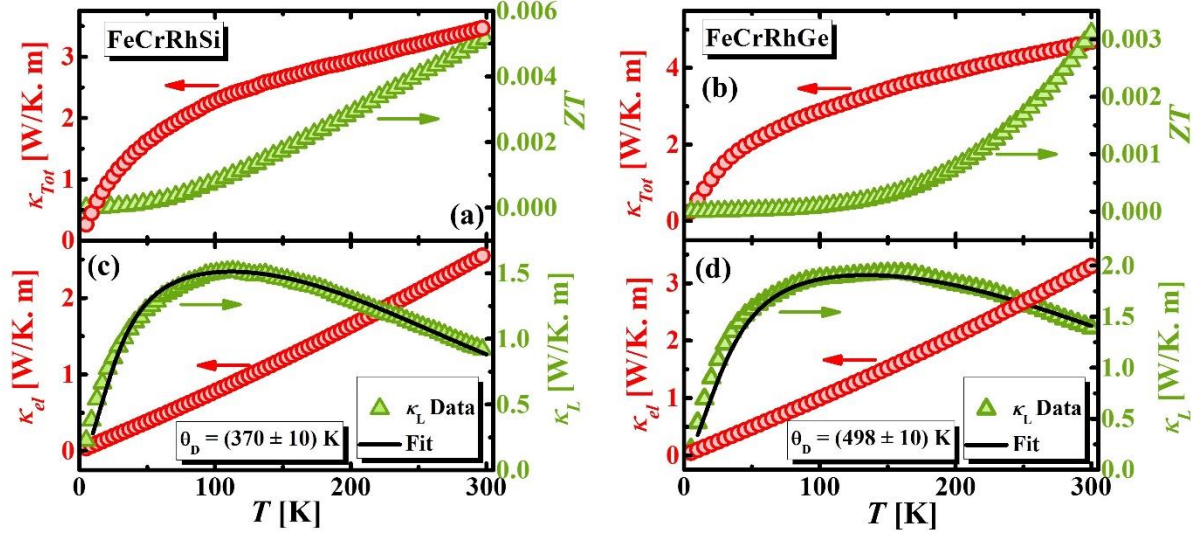


Figure 4. Temperature dependence of the total thermal conductivity, κ_{Tot} (left vertical scale) and thermoelectric figure of merit, ZT (right vertical scale) for (a) FeCrRhSi and (b) FeCrRhGe. Temperature dependence of the electronic thermal conductivity, κ_{el} (left vertical scale) and lattice thermal conductivity, κ_L (right vertical scale) for (c) FeCrRhSi and (d) FeCrRhGe.

Generally, while the Umklapp scattering process is crucial at high temperatures, the phonon-boundary scattering plays a dominating role at low temperatures. [86] On the contrary, point-defect scattering becomes a dominating factor in the intermediate temperature regime and hence, strongly determines the shape and position of the phonon peak/maximum. [86] The point defect scattering has two main contributions: mass fluctuations and strain field scatterings. In other words, significant differences in atomic masses, atomic radii and, interatomic coupling forces between the constituent atoms, or, between the host and impurity atoms can strongly influence the relaxation time for point defect scattering. [82] It is known that Ge has ≈ 2.6 times higher atomic mass and nearly ≈ 1.04 times larger atomic radius than Si. Therefore, a significant difference in the relaxation time for point defect scattering is expected between FeCrRhSi and FeCrRhGe. From our fitting we observed that the parameter A is nearly 4 times larger in FeCrRhGe than in FeCrRhSi. In another Heusler system, $\text{Fe}_2\text{Va}_{1-x}\text{Si}_x$, the parameter

A is reported to increase monotonically with increasing Si doping, where Si has slightly higher atomic mass than Al. [86] It was also mentioned that not only the atomic mass fluctuations but also lattice imperfections introduced by Si doping can play an important role towards the enhanced point defect scattering. However, for a more concrete explanation for the large difference in the parameter A between FeCrRhSi and FeCrRhGe, a rigorous theoretical calculation is needed to estimate the individual contribution of the mass fluctuations and strain field scattering contributions. On the other hand, the observed difference in the parameter B between the FeCrRhSi and FeCrRhGe alloys can be due to the fact that the Umklapp scattering process plays a dominating role at high temperatures whereas the measurement/fitting was limited to $T \ll \theta_D$ for FeCrRhGe. Since FeCrRhGe has higher θ_D than FeCrRhSi, a more accurate value of the parameter B could be obtained if the measurement/fitting limit could have been extended to higher temperatures ($T \geq \theta_D$).

3.5. Transverse magneto-thermoelectric transport: Anomalous Nernst effect

Next, we discuss the transverse magneto-thermoelectric transport properties of FeCrRhSi and FeCrRhGe. The schematic illustration of the experimental geometry used for these measurements is shown in **Fig. 5(a)**. [87] In presence of the temperature gradient applied along the thickness of the sample, (y -direction), a temperature difference, $\Delta T = (T_{Hot} - T_{Cold})$ between the bottom (hot) and top (cold) copper plates is generated, where, T_{Hot} and T_{Cold} are the temperatures of the hot and cold copper plates, respectively. Because of this temperature difference between the bottom and top copper plates, the sample surface which is in contact with the bottom copper plate will have a higher temperature than the surface that is in contact with the top copper plate. In presence of this temperature difference between the opposite surfaces of the sample along its thickness, and an external DC magnetic field applied

along the x -direction, a transverse anomalous Nernst voltage is induced in the sample along the z -direction.

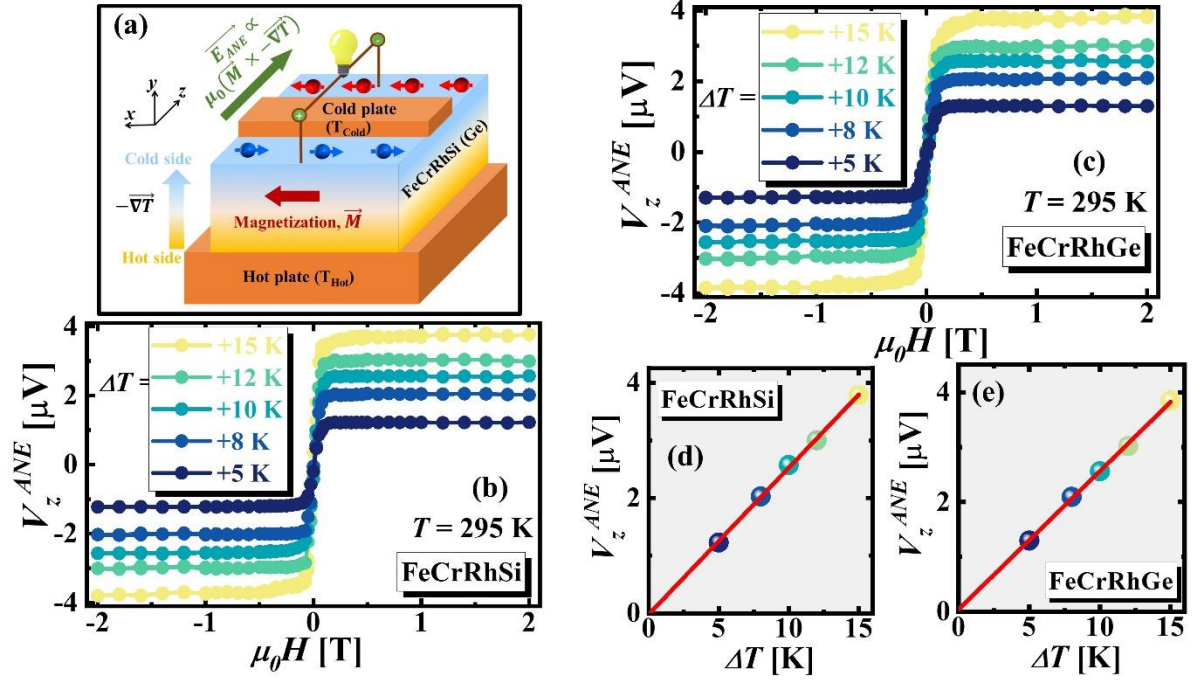


Figure 5. (a) Schematic illustration of the anomalous Nernst measurement configuration using our spin caloritronic set up. Magnetic field dependence of the anomalous Nernst voltage, $V_z^{ANE}(H)$ for different values of ΔT in the range: $+5 \text{ K} \leq \Delta T \leq +15 \text{ K}$ at $T = 295 \text{ K}$ for (b) FeCrRhSi and (c) FeCrRhGe. The background corrected anomalous Nernst voltage, $V_z^{ANE}(\mu_0 H_{sat}) = \left[\frac{V_z^{ANE}(+\mu_0 H_{sat}) - V_z^{ANE}(-\mu_0 H_{sat})}{2} \right]$ as a function of ΔT are shown for (d) FeCrRhSi and (e) FeCrRhGe.

Figures. 5(b) and (c) demonstrate the magnetic field dependence ($\mu_0 H$) of the anomalous Nernst voltage, $V_z^{ANE}(\mu_0 H)$ for different values of ΔT in the range: $+5 \text{ K} \leq \Delta T \leq +15 \text{ K}$ for the FeCrRhSi and FeCrRhGe, respectively at a fixed average sample temperature $T = \frac{T_{Hot} + T_{Cold}}{2} = 295 \text{ K}$. Clearly, the $V_z^{ANE}(H)$ signal strength increases with increasing ΔT for both the samples, which is in agreement with the fact that the transverse electric field induced by the ANE ($\vec{E}_z^{ANE}$) is related to the applied temperature gradient ($\vec{\nabla T}$) through the

expression, [13] $\overrightarrow{E_z^{ANE}} \propto (\mu_0 \overrightarrow{M} \times -\nabla T)$, where $\overrightarrow{M}$ is the magnetization vector of the sample under consideration. **Figs. 5(d) and (e)** show the ΔT -dependence of the background corrected antisymmetrized anomalous Nernst voltage, $V_z^{ANE}(\mu_0 H_{sat}) = \left[\frac{V_z^{ANE}(+\mu_0 H_{sat}) - V_z^{ANE}(-\mu_0 H_{sat})}{2} \right]$ for the FeCrRhSi and FeCrRhGe samples, respectively at $T = 295$ K, where $\mu_0 H = \mu_0 H_{sat}$ is the saturation magnetic field. It is evident that $V_z^{ANE}(\mu_0 H_{sat})$ for both the samples varies linearly with ΔT , indicating intrinsic contribution of the thermally induced ANE in FeCrRhSi and FeCrRhGe [23,27].

To understand the temperature evolution of the ANE and its underlying mechanism in these samples, we show the $V_z^{ANE}(H)$ curves for FeCrRhSi and FeCrRhGe in **Fig. 6(a) and Fig. 6(b)**, respectively at selected average sample temperatures in the range $130 \text{ K} \leq T \leq 295 \text{ K}$ for a fixed value of $\Delta T = +15 \text{ K}$. It is evident from both **Fig. 6(a) and Fig. 6(b)** that $V_z^{ANE}(H)$ signal strength for both FeCrRhSi and FeCrRhGe alloys decreases with decreasing temperature. To have a better visualization of the temperature evolution of the ANE signal in FeCrRhSi and FeCrRhGe, the estimation of the anomalous Nernst coefficient, S_{zy}^{ANE} (defined as, [25,88] $S_{zy}^{ANE} = [E_z^{ANE} / (\nabla_y T)]$) at each temperature is required. S_{zy}^{ANE} at a given temperature can be accurately estimated using the expression, [9,23,27] $S_{zy}^{ANE}(\mu_0 H_{sat}, T) = \left(\frac{V_z^{ANE}(\mu_0 H_{sat})}{\Delta T_{eff}} \right) \times \left(\frac{L_y}{L_z} \right) = \left[\frac{V_z^{ANE}(+\mu_0 H_{sat}) - V_z^{ANE}(-\mu_0 H_{sat})}{2 \Delta T_{eff}} \right] \times \left(\frac{L_y}{L_z} \right)$. Here, L_y ($\approx 1.2 \text{ mm}$ for FeCrRhSi and $\approx 1 \text{ mm}$ for FeCrRhGe) is the thickness of the sample, L_z ($\approx 3 \text{ mm}$) is the distance between the voltage probes for the ANE measurements and, $\Delta T_{eff} = (T_1 - T_2)$ represents the effective temperature difference between the two opposite isothermal surfaces of the sample, one of which is in contact with the hot copper plate and the other one with the cold copper plate, as

shown in **Fig. 7(a)**. Here, T_1 (T_2) is the temperature of the sample surface which is in contact with the hot (cold) copper plate.

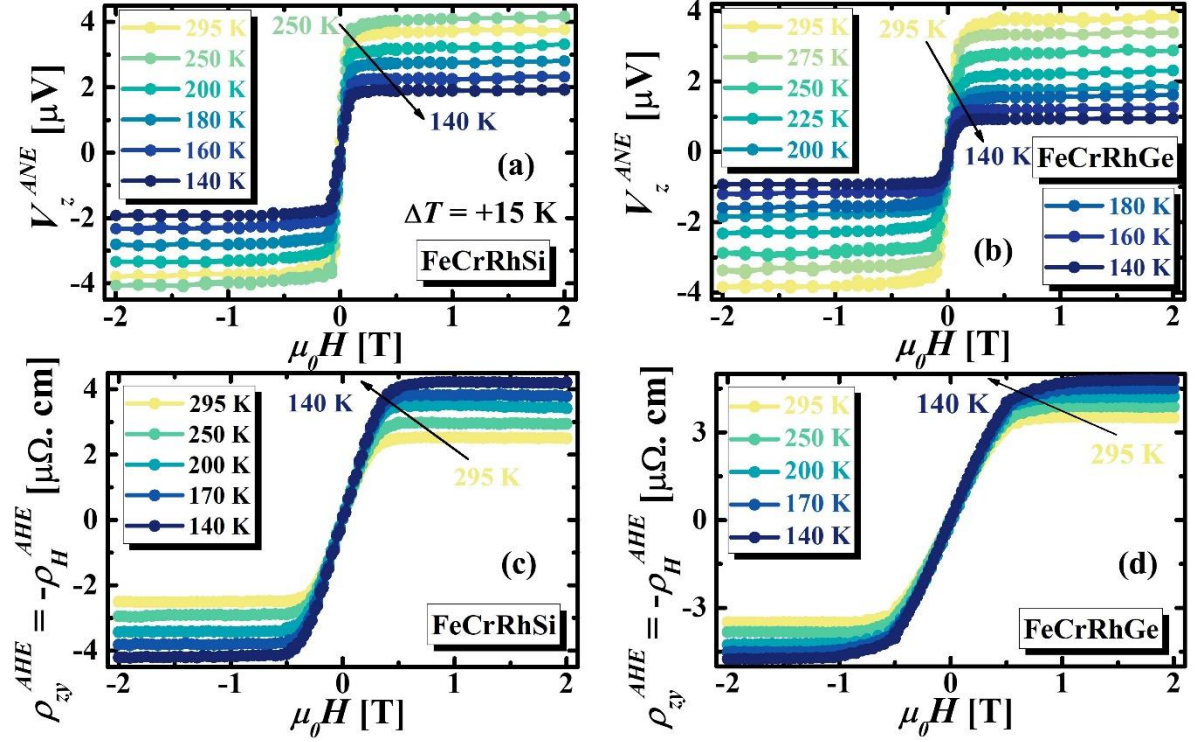


Figure 6. Magnetic field dependence of the anomalous Nernst voltage, $V_z^{ANE}(H)$ for (a) FeCrRhSi and (b) FeCrRhGe, at selected temperatures in the range $140 \text{ K} \leq T \leq 295 \text{ K}$ for fixed $\Delta T = +15 \text{ K}$. Magnetic field dependence of anomalous Hall resistivity $\rho_{zy}^{AHE}(H)$ for (c) FeCrRhSi and (d) FeCrRhGe at selected temperatures in the range: $140 \text{ K} \leq T \leq 295 \text{ K}$.

Note that the applied thermal gradient is dropped not only across the samples under consideration, but also across the N-grease layers on both sides of the samples, at the interface between the hot/cold copper plates and the N-grease layers as well as at the interface between the sample surfaces and the N-grease layers, and hence $\Delta T_{eff} \neq \Delta T$. Therefore, for an accurate determination of S_{zy}^{ANE} , a careful estimation of ΔT_{eff} is of utmost importance. According to **Fig. 7(a)**, the total thermal resistance across the entire system can be written as,

$$R_T = \left(\frac{L_{\text{FeCrRhX}}}{\kappa_{\text{FeCrRhX}}} + \frac{2L_{\text{N-Grease}}}{\kappa_{\text{N-Grease}}} + 2R_{int}^{Cu-N} + 2R_{int}^{\text{FeCrRhX-N}} \right), \text{ where } L_{\text{FeCrRhX}} \text{ and } \kappa_{\text{FeCrRhX}} \text{ are}$$

the thickness and the thermal conductivity of the FeCrRhX ($X = \text{Si, Ge}$) alloys, and $L_{\text{N-Grease}}$ and $\kappa_{\text{N-Grease}}$ are the thickness and thermal conductivity of the N-grease layers, R_{int}^{Cu-N} is the interfacial thermal resistance between the N-grease and hot/cold copper plates, and $R_{int}^{FeCrRhX-N}$ is the interfacial thermal resistance between the N-grease and the FeCrRhX samples. Note that the purpose of using the Apiezon N grease is to reduce interfacial thermal resistance between the sample surface and hot/cold copper plates, as the Apiezon N grease is regarded very efficient thermal bonding agent compared to other Si or copper based thermal bonding agents. [89] As seen in **Fig. 7(b)**, $\kappa_{\text{N-Grease}}$ is comparable to κ_{FeCrRhX} in contrast to the large difference in thermal conductivities between copper ($\approx 400 \text{ W/(m. K)}$ at room temperature) and the Apiezon N grease ($\approx 0.25 \text{ W/(m. K)}$ at room temperature) as well as the FeCrRhX alloys ($\approx 3 - 4.5 \text{ W/(m. K)}$ at room temperature). Hence, $R_{int}^{FeCrRhX-N}$ is very small compared to R_{int}^{Cu-N} . Furthermore, interfacial thermal resistance is known to decrease with decreasing the mean surface roughness. [90] Since our sample surfaces are highly polished, the interfacial thermal resistance between the sample surfaces and N-grease is reduced further. Additionally, the interfacial thermal resistance is reported to decrease with increasing the applied contact pressure. [90] In our ANE measurement set up, the samples are tightly sandwiched between the hot/cold copper plates by tightening the Mo screws which apply sufficient contact pressure at the interfaces between sample surfaces and N-grease. Therefore, we believe that the interfacial thermal resistance between the Apiezon N-grease and sample surfaces is negligible for our ANE measurements, and hence, $R_{int}^{FeCrRhX-N} \approx 0$. Therefore, considering the temperature drop across the sample, the N-grease layers and at the interface between the hot/cold copper plates and N-grease layers, the total thermal resistance becomes, $R_T = \left(\frac{L_{\text{FeCrRhX}}}{\kappa_{\text{FeCrRhX}}} + \frac{2L_{\text{N-Grease}}}{\kappa_{\text{N-Grease}}} + 2R_{int}^{Cu-N} \right)$. In the steady state, the heat flux is constant throughout the system, and we can write, $\frac{\Delta T}{R_T} = \kappa_{\text{FeCrRhX}} \left(\frac{\Delta T_{eff}}{L_{\text{FeCrRhX}}} \right)$. Alternatively, ΔT_{eff} can be expressed as,

$$\Delta T_{eff} = \frac{\Delta T}{\left[1 + \left(2R_{int}^{Cu-N} + \left(\frac{2L_{N-Grease}}{\kappa_{N-Grease}}\right)\right) \left(\frac{\kappa_{FeCrRhX}}{L_{FeCrRhX}}\right)\right]}$$
 The interfacial thermal conductance between Cu-foil and Apiezon N-grease for less than 1kPa applied pressure is reported to be around 10,000 W/(m².K) in the temperature range 100 K $\leq T \leq$ 300 K, [91] indicating an interfacial thermal resistance of $R_{int}^{Cu-N} \approx 1 \times 10^{-4}$ (m².K)/W. Using the reported values of the temperature dependence of $\kappa_{N-Grease}$, [92] and considering $L_{N-Grease} \approx 10$ μ m, [26] $L_{FeCrRhX} = L_y \approx 1.2$ mm for FeCrRhSi and ≈ 1 mm for FeCrRhGe, we determined the temperature dependence of ΔT_{eff} for both the samples.

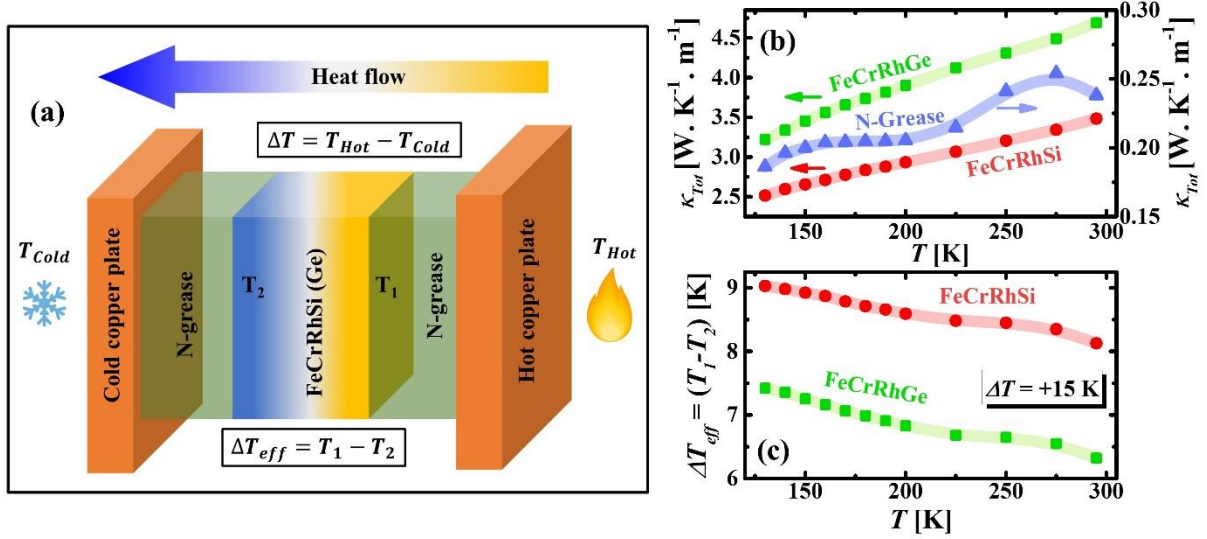


Figure 7. (a) Heat flow and the temperature gradient across the sample. (b) Temperature dependences of the total thermal conductivities, $\kappa_{N-Grease}(T)$, $\kappa_{FeCrRhSi}(T)$ and $\kappa_{FeCrRhGe}(T)$. (c) Temperature dependence of the effective temperature gradient $\Delta T_{eff}(T)$ for FeCrRhSi and FeCrRhGe for a fixed $\Delta T = +15$ K.

Figure 7(b) displays the temperature dependences of $\kappa_{N-Grease}$ and $\kappa_{FeCrRhX}$, where $\kappa_{N-Grease}(T)$ was obtained by interpolating the reported values of the temperature dependence of $\kappa_{N-Grease}$ [92]. The estimated temperature profiles of ΔT_{eff} for the FeCrRhSi and FeCrRhGe samples are shown in **Fig. 7(c)**. Using the estimated values of $\Delta T_{eff}(T)$, we have

determined the temperature dependences of the background-corrected antisymmetrized anomalous Nernst coefficient (S_{zy}^{ANE}) which was previously defined as, [9,23,27]

$$S_{zy}^{ANE}(\mu_0 H_{sat}, T) = \left[\frac{V_z^{ANE}(+\mu_0 H_{sat}) - V_z^{ANE}(-\mu_0 H_{sat})}{2\Delta T_{eff}} \right] \times \left(\frac{L_y}{L_z} \right),$$

as demonstrated in **Figs. 8(a)** and **8(b)** for the FeCrRhSi and FeCrRhGe samples, respectively. Here, the estimated values of S_{zy}^{ANE} for the FeCrRhSi and FeCrRhGe samples at $T = 295$ K are $(0.272 \pm 0.02) \mu\text{V} \cdot \text{K}^{-1}$ and, $(0.295 \pm 0.02) \mu\text{V} \cdot \text{K}^{-1}$, respectively. To the best of our knowledge, these values of S_{zy}^{ANE} for the FeCrRhSi and FeCrRhGe samples are the highest among all the other EQHAs, *e.g.*, CrRuCoGe ($\approx 0.058 \mu\text{V} \cdot \text{K}^{-1}$), [26] the semi-metallic Heusler alloy CoFeVSb ($\approx 0.039 \mu\text{V} \cdot \text{K}^{-1}$), [20] the spin gapless semiconductor CoFeCrGa ($\approx 0.018 \mu\text{V} \cdot \text{K}^{-1}$), [12] *etc.* A more detailed comparison with different Heusler alloys and a range of other magnetic materials is discussed later. As seen in **Fig. 8(a)**, while S_{zy}^{ANE} for the FeCrRhSi sample first increases slowly with decreasing temperature in the range: $250 \text{ K} \leq T \leq 295 \text{ K}$, it shows a slope change around 250 K which is followed by a monotonic decrease below 250 K down to the lowest measured temperature. On the other hand, S_{zy}^{ANE} for the FeCrRhGe sample decreases monotonically with decreasing temperature throughout the measured temperature range, as displayed in **Fig. 8(b)**.

3.6. Discussion

Now let us give some insight into the origins of the observed behavior of $S_{zy}^{ANE}(\mu_0 H_{sat}, T)$ in the two samples, for which we need to understand the correlation between S_{zy}^{ANE} and other fundamental transport parameters. Thermoelectric effects can be understood from the general expression of charge current $\vec{J}$ flowing under the influence of an electric field $\vec{E}$ in the presence of an external temperature gradient $\vec{\nabla T}$ as, $\vec{J} = \tilde{\sigma} \cdot \vec{E} + \tilde{\alpha} \cdot (-\vec{\nabla T})$, where $\tilde{\sigma}$ and $\tilde{\alpha}$ are the electrical conductivity tensor and thermoelectric conductivity tensor [25,27,31].

Without loss of generality and considering open circuit conditions, *i.e.*, $J = 0$, if the temperature gradient is applied along the y direction and the external magnetic field is applied along the x -direction, in accordance with the right-handed Cartesian coordinate system as presented in **Fig. 5(a)**, the transverse component of the electric field can be expressed as, $E_z =$

$$\left[\left(\frac{\sigma_{yy}\alpha_{zy} - \sigma_{zy}\alpha_{yy}}{\sigma_{yy}^2 + \sigma_{zy}^2} \right) \cdot \nabla_y T \right]. [25,27,31] \text{ In general, } \sigma_{yy}^2 \gg \sigma_{zy}^2, \text{ and } \rho_{zy} = -\rho_{yz}, \text{ and } \sigma_{zy} = -\sigma_{yz}.$$

Here, $\rho_{yz}(\sigma_{yz})$ and $\rho_{yy}(\sigma_{yy})$ are Hall resistivity (conductivity) and longitudinal resistivity (conductivity) components. Therefore, the transverse electric field becomes, $E_z = [(\rho_{yy}\alpha_{zy} + \rho_{zy}\alpha_{yy}) \cdot \nabla_y T]$, where $\rho_{yy} = (1/\sigma_{yy})$ and $\rho_{zy} = -\left[\frac{\sigma_{zy}}{\sigma_{yy}^2 + \sigma_{zy}^2} \right] \approx \left[\frac{-\sigma_{zy}}{\sigma_{yy}^2} \right]$. [25,27,31] Therefore,

considering anomalous components of α_{zy} and ρ_{zy} , and using the definition of anomalous Nernst coefficient, S_{zy}^{ANE} can be expressed as, [25,31,93] $S_{zy}^{ANE} = [E_z^{ANE}/(\nabla_y T)] =$

$(\rho_{yy}\alpha_{zy}^A + \rho_{zy}^{AHE}\alpha_{yy}) = \left[\rho_{yy}\alpha_{zy}^A + \left(\frac{\rho_{zy}^{AHE}}{\rho_{yy}} \right) S_{yy} \right]$; where, α_{zy}^A is the anomalous off-diagonal transverse thermoelectric conductivity (Peltier conductivity), ρ_{zy}^{AHE} is the anomalous Hall resistivity, and $S_{yy} = \rho_{yy}\alpha_{yy}$ is the longitudinal Seebeck coefficient in accordance with the right-handed Cartesian coordinate system as presented in **Fig. 5(a)**. Now, the resistivity tensor

in three dimensions can be written as, [88] $\tilde{\rho} = \begin{bmatrix} \rho_{xx} & \rho_{xy} & \rho_{xz} \\ \rho_{yx} & \rho_{yy} & \rho_{yz} \\ \rho_{zx} & \rho_{zy} & \rho_{zz} \end{bmatrix}$. For an isotropic

polycrystalline material, it is reasonable to assume that $\rho_{xx} = \rho_{yy} = \rho_{zz} = \rho =$ isotropic longitudinal resistivity and, $\rho_{xy} = \rho_{yz} = \rho_{zx} = \rho_H =$ isotropic Hall resistivity.

Here ρ_{xy} , ρ_{yz} and ρ_{zx} are the Hall resistivity when the magnetic field is applied along the z , x and y directions, respectively. Since, in the present study, the external magnetic field is applied along the x -direction, $\rho_{xy} = \rho_{zx} = 0$, and $\rho_{yz} = \rho_H$. For such isotropic polycrystalline materials, we can also consider that $S_{xx} = S_{yy} = S_{zz} = S =$

isotropic longitudinal Seebeck coefficient. Considering all these approximations, the

anomalous Nernst coefficient for our polycrystalline FeCrRhSi and FeCrRhGe alloys can be

expressed as, [25,93] $S_{zy}^{ANE} = \left[\rho_{yy} \alpha_{zy}^A + \left(\frac{\rho_{zy}^{AHE}}{\rho_{yy}} \right) S_{yy} \right] = \left[\rho \cdot \alpha_{zy}^A - S \cdot \left(\frac{\rho_H^{AHE}}{\rho} \right) \right] = \left[\rho \cdot \alpha_{zy}^A - \right.$

$S \cdot \tan \theta_H \left. \right]$, where $\rho_H^{AHE} = \rho_{yz}^{AHE} = -\rho_{zy}^{AHE}$ is the isotropic anomalous Hall resistivity and

$\tan \theta_H = \left(\frac{\rho_H^{AHE}}{\rho} \right)$ is the anomalous Hall angle. It is therefore evident that S_{zy}^{ANE} can be regarded

as a linear combination of two primary terms. The first term $S_1^{ANE} = \rho \cdot \alpha_{zy}^A$ represents the

transverse thermoelectric conduction that directly transforms the thermal gradient $\nabla_y \mathbf{T}$ into

transverse electric current density, $\mathbf{J}_z^A = -\alpha_{zy}^A \cdot \nabla_y \mathbf{T}$. [25,93,94] On the other hand, the second

term $S_2^{ANE} = -S \cdot \tan \theta_H$ indicates that the thermally generated charge carrier flow induced by

the longitudinal thermoelectric effect is transformed into a transverse flow of carriers as a result

of the anomalous Hall effect (AHE). [93,94] Note that, it is also convenient to express the

anomalous Hall conductivity (σ_H^{AHE}) and the longitudinal electrical conductivity (σ) as, [2,7,9]

$\sigma_H^{AHE} = \left[\frac{-\rho_H^{AHE}}{(\rho)^2 + (\rho_H^{AHE})^2} \right] \approx \left[\frac{-\rho_H^{AHE}}{(\rho)^2} \right]$ and $\sigma = \left[\frac{\rho}{(\rho)^2 + (\rho_H^{AHE})^2} \right] \approx \left[\frac{1}{\rho} \right]$, respectively, considering

$(\rho)^2 \gg (\rho_H^{AHE})^2$. Therefore, S_{zy}^{ANE} can be alternatively expressed as, $S_{zy}^{ANE} = \left[\rho_{yy} \alpha_{zy}^A + \right.$

$\left. \left(\frac{\rho_{zy}^{AHE}}{\rho_{yy}} \right) S_{yy} \right] = \left[\rho \cdot \alpha_{zy}^A - S \cdot \left(\frac{\rho_H^{AHE}}{\rho} \right) \right] = \left[\rho \cdot \alpha_{zy}^A + S \cdot \rho \left\{ \frac{-\rho_H^{AHE}}{(\rho)^2} \right\} \right] = \rho \cdot \left[\alpha_{zy}^A + S \cdot \sigma_H^{AHE} \right]$.

According to the Mott's relations, α_{zy}^A and S can be expressed as energy derivative of σ_H^{AHE}

and σ at the E_F as, [25] $\left(\frac{\alpha_{zy}^A}{T} \right) = \frac{\pi^2 k_B^2}{3e} \left(\frac{\partial \sigma_{zy}^{AHE}}{\partial E} \right)_{E=E_F}$ and, $\left(\frac{S}{T} \right) = \frac{\pi^2 k_B^2}{3e\sigma} \left(\frac{\partial \sigma}{\partial E} \right)_{E=E_F}$, respectively.

Now, according to the power law for AHE, ρ_{zy}^{AHE} depends on ρ through the expression:

$\rho_{zy}^{AHE} = -\rho_{yz}^{AHE} = -\rho_H^{AHE} = -\lambda \rho^n$, where λ represents the spin-orbit coupling constant and n

is a constant exponent that determines the nature of the underlying mechanism for AHE and

ANE. When $n \approx 1$, the extrinsic skew scattering is the dominant mechanism, and when $n \approx$

2, the intrinsic Berry curvature or, the side jump is the dominating mechanism for the

anomalous Nernst/Hall transport [6]. Therefore, incorporating the power law for AHE, the

expressions for α_{zy}^A and S can be written as, $\left(\frac{\alpha_{zy}^A}{T}\right) = \frac{\pi^2 k_B^2}{3e} \left(\frac{\partial \sigma_{zy}^{AHE}}{\partial E}\right)_{E=E_F} =$
 $-\frac{\pi^2 k_B^2}{3e} \left(\frac{\partial \sigma_{yz}^{AHE}}{\partial E}\right)_{E=E_F} = -\frac{\pi^2 k_B^2}{3e} \left(\frac{\partial \sigma_H^{AHE}}{\partial E}\right)_{E=E_F} = -\frac{\pi^2 k_B^2}{3e} \left[\frac{\partial}{\partial E} \left\{\frac{-\rho_H^{AHE}}{(\rho)^2}\right\}\right]_{E=E_F} =$
 $-\frac{\pi^2 k_B^2}{3e} \left[\frac{\partial}{\partial E} \{-\lambda \rho^{n-2}\}\right]_{E=E_F}$ and, $\left(\frac{S}{T}\right) = -\frac{\pi^2 k_B^2}{3e\rho} \left(\frac{\partial \rho}{\partial E}\right)_{E=E_F}$. Considering all the above mentioned expressions, $S_{zy}^{ANE}(T)$ can be transformed into the following equation, [2,27]

$$S_{zy}^{ANE}(T) = \rho^{n-1} \left[\frac{\pi^2 k_B^2 T}{3e} \left(\frac{\partial \lambda}{\partial E}\right)_{E=E_F} - (n-1)\lambda \cdot S \right] \quad (2)$$

We fitted the $S_{zy}^{ANE}(T)$ data for FeCrRhSi and FeCrRhGe within the measured temperature range ($140 \text{ K} \leq T \leq 250 \text{ K}$ for the FeCrRhSi sample) using **Eqn. (2)** and considering λ , $\left(\frac{\partial \lambda}{\partial E}\right)_{E=E_F}$, and n as the fitting parameters. The best fits were achieved for $n = (1.982 \pm 0.4)$ and $n = (1.866 \pm 0.2)$ for FeCrRhSi and FeCrRhGe, respectively implying that the origin of ANE in both these samples is dominated either by the intrinsic Berry curvature or, the side jump mechanism in the measured temperature regime [6,12]. It is noteworthy that the previous reports on FeCrRhSi and FeCrRhGe [44,45] observed that AHE in both these samples is dominated by the Berry curvature, which was further supported by the first principle calculations. Since AHE and ANE share the common origin, [7] it is reasonable to conclude that the observed ANE in both FeCrRhSi and FeCrRhGe samples is dominated by the Berry curvature.

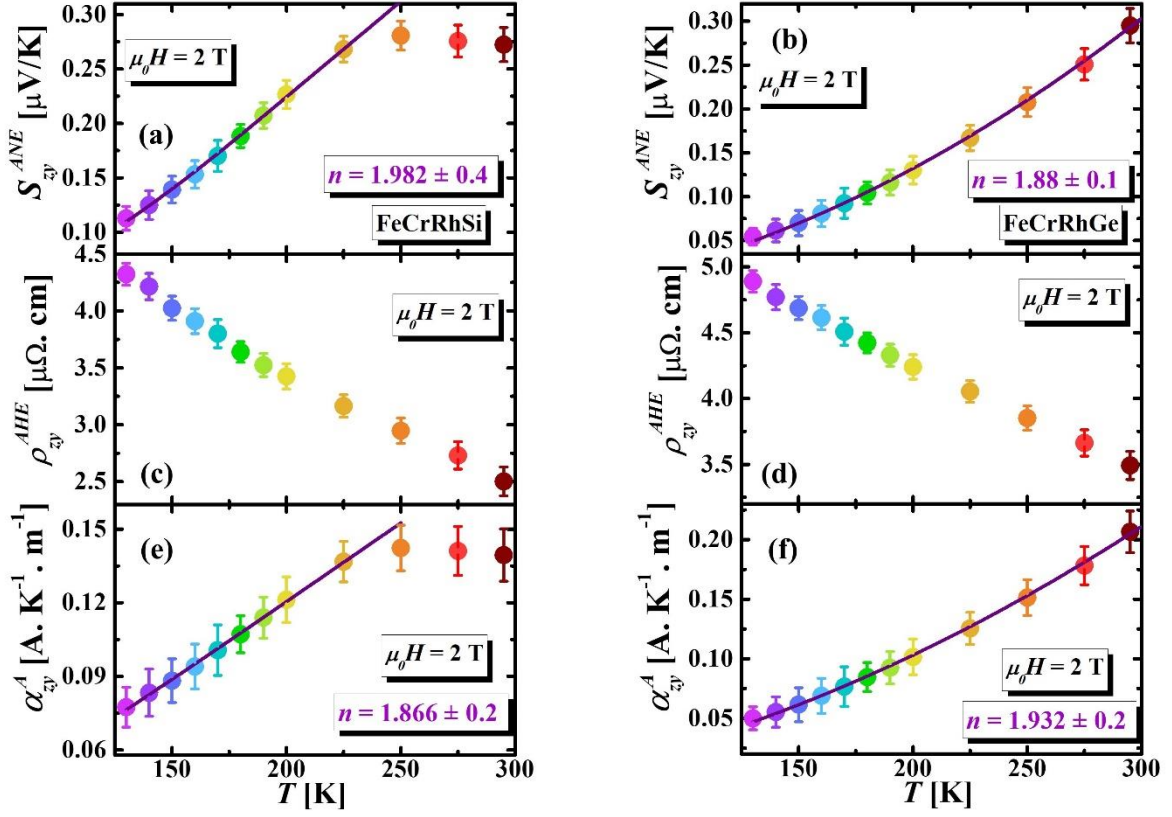


Figure 8. Temperature dependence of the background-corrected anomalous Nernst coefficient,

$$S_{zy}^{ANE}(\mu_0 H_{sat}, T) = \left[\frac{V_z^{ANE}(+\mu_0 H_{sat}) - V_z^{ANE}(-\mu_0 H_{sat})}{2\Delta T_{eff}} \right] \times \left(\frac{L_y}{L_z} \right), \text{ anomalous Hall resistivity,}$$

$\rho_{zy}^{AHE}(\mu_0 H_{sat}, T)$ and anomalous off-diagonal thermoelectric conductivity, $\alpha_{zy}^A(T)$ for (a,c,e)

FeCrRhSi and (b,d,f) FeCrRhGe.

To further support our conclusion, it is imperative to determine and analyze the $\alpha_{zy}^A(T)$ data. Using the expression, $S_{zy}^{ANE} = \left[\rho_{yy} \alpha_{zy}^A + \left(\frac{\rho_{zy}^{AHE}}{\rho_{yy}} \right) S_{yy} \right]$, we can write α_{zy}^A as, [93] $\alpha_{zy}^A = \left(\frac{1}{\rho_{yy}} \right) \cdot \left[S_{zy}^{ANE} - \left(\frac{\rho_{zy}^{AHE}}{\rho_{yy}} \right) S_{yy} \right] = \left[\sigma \cdot S_{zy}^{ANE} - S \cdot \left(-\frac{\rho_H^{AHE}}{\rho^2} \right) \right] = (\sigma \cdot S_{zy}^{ANE} - S \cdot \sigma_H^{AHE})$. Therefore, following the Mott's relations, α_{zy}^A can be expressed as, [2,14,27]

$$\alpha_{zy}^A(T) = \rho^{n-2} \left[\frac{\pi^2 k_B^2 T}{3e} \left(\frac{\partial \lambda}{\partial E} \right)_{E=E_F} - (n-2) \lambda \cdot S \right] \quad (3)$$

In order to estimate $\alpha_{zy}^A(T)$, it is important to have experimental data for $\sigma_{zy}^{AHE}(T)$ and hence $\rho_{zy}^{AHE}(T)$. We have performed AHE measurements on both the FeCrRhSi and FeCrRhGe samples. **Figures 6(c) and (d)** display the magnetic field dependence of anomalous Hall resistivity $\rho_{zy}^{AHE}(\mu_0 H) = -\rho_{yz}^{AHE}(\mu_0 H) = -\rho_H^{AHE}(\mu_0 H)$ for the FeCrRhSi and FeCrRhGe samples, respectively at selected temperatures in the range: $130\text{K} \leq T \leq 295\text{ K}$. It is evident that unlike $V_z^{ANE}(H)$, $|\rho_H^{AHE}(H)|$ increases with decreasing temperature for both FeCrRhSi and FeCrRhGe. **Figures 8(c) and (d)** display the temperature dependence of the anti-symmetrized anomalous Hall resistivity, $\rho_{zy}^{AHE}(\mu_0 H_{sat}, T) = -\rho_H^{AHE}(\mu_0 H_{sat}, T) = \frac{1}{2} [\{-\rho_H^{AHE}(+\mu_0 H_{sat}, T)\} - \{-\rho_H^{AHE}(-\mu_0 H_{sat}, T)\}]$ for the FeCrRhSi and FeCrRhGe samples, respectively. Evidently, $|\rho_{zy}^{AHE}(\mu_0 H_{sat}, T)|$ for both these samples increases monotonically with decreasing temperature throughout the measured temperature range. In this context, it is noteworthy that both $S_{zy}^{ANE}(T)$ and $|\rho_{zy}^{AHE}(\mu_0 H_{sat}, T)|$ are expected show a peak position/slope change where the $M(T)$ shows a slope change. However, it was observed that $\rho_{zy}^{AHE}(\mu_0 H_{sat}, T)$ (and hence $\sigma_{zy}^{AHE}(\mu_0 H_{sat}, T)$) replicates the $M(T)$ behavior and shows slope changes at the onset of ferromagnetic transition (T_C). [9] On the other hand, S_{zy}^{ANE} usually does not show a slope change/maximum at the T_C , but increases rapidly around the T_C , shows a maximum value at a certain temperature below the T_C , and then decreases towards zero at low temperatures. [9,24] This is why, S_{zy}^{ANE} and ρ_{zy}^{AHE} do not always show maximum/slope change at the same temperature [19], possibly because S_{zy}^{ANE} is composed of two terms as, $S_{zy}^{ANE} = \left[\rho \cdot \alpha_{zy}^A + \left(\frac{\rho_{zy}^{AHE}}{\rho} \right) S \right]$, and hence, S_{zy}^{ANE} depends not only on ρ_{zy}^{AHE} (and hence σ_{zy}^{AHE}) but also on other fundamental parameters such as, ρ , S and α_{zy}^A [14]. Since T_C of FeCrRhSi is nearly 100 K lower than FeCrRhGe, the slope change/maximum in $S_{zy}^{ANE}(T)$ is expected to occur at lower temperatures in FeCrRhSi than in FeCrRhGe. Nevertheless, the maximum/slope change in

$S_{zy}^{ANE}(T)$ is expected to appear below $T_C = 458$ and 550 K in FeCrRhSi and FeCrRhGe, respectively. As shown in **Figs. 2(a)** and **(b)**, while both ZFCW and FCW $M(T)$ continue to increase with increasing temperature up to 390 K for FeCrRhGe, those for FeCrRhSi first increase with temperature up to 300 K, exhibit a broad maximum in the vicinity of 300 K and then tend to decrease slowly with further increasing temperature which is followed by an abrupt drop above 350 K, which is indicative of the starting point of ferromagnetic to paramagnetic transition. Therefore, the slope-change in S_{zy}^{ANE} for FeCrRhSi around 250 K is associated with the occurrence of broad maximum in the ZFCW and FCW $M(T)$ in the temperature range: $300 \text{ K} \leq T \leq 350 \text{ K}$ as a result of the ferromagnetic transition. Since the ANE measurements were performed in the temperature range $130 \text{ K} \leq T \leq 295 \text{ K}$, we could only detect the slope change in $S_{zy}^{ANE}(T)$ for the FeCrRhSi sample (around 250 K) and possibly the slope change/maximum in $S_{zy}^{ANE}(T)$ for FeCrRhGe occurs at $T \gg 295 \text{ K}$, which lies beyond the measured temperature range. Also, based on our previous argument, since the slope change in ρ_{zy}^{AHE} occurs at the onset of ferromagnetic transition, which also lies much above our measured temperature range ($130 \text{ K} \leq T \leq 295 \text{ K}$), no slope change was observed in $\rho_{zy}^{AHE}(T)$ for either of the samples within the measured temperature range. As shown in **Figs. 8(e)** and **(f)**, the estimated $\alpha_{zy}^A(T)$ for both FeCrRhSi and FeCrRhGe mimics the behavior of the corresponding $S_{zy}^{ANE}(T)$. We fitted the $\alpha_{zy}^A(T)$ data for both the FeCrRhSi and FeCrRhGe samples using **Eqn. (3)**, and the best fits were obtained for $n = (1.88 \pm 0.1)$ and $n = (1.932 \pm 0.2)$ for FeCrRhSi and FeCrRhGe. Clearly, the values of the exponent n obtained from the $\alpha_{zy}^A(T)$ fits are close to those obtained by fitting the $S_{zy}^{ANE}(T)$ data for both the samples and hence, confirms that the observed ANE in FeCrRhSi and FeCrRhGe is dominated by the Berry curvature [6].

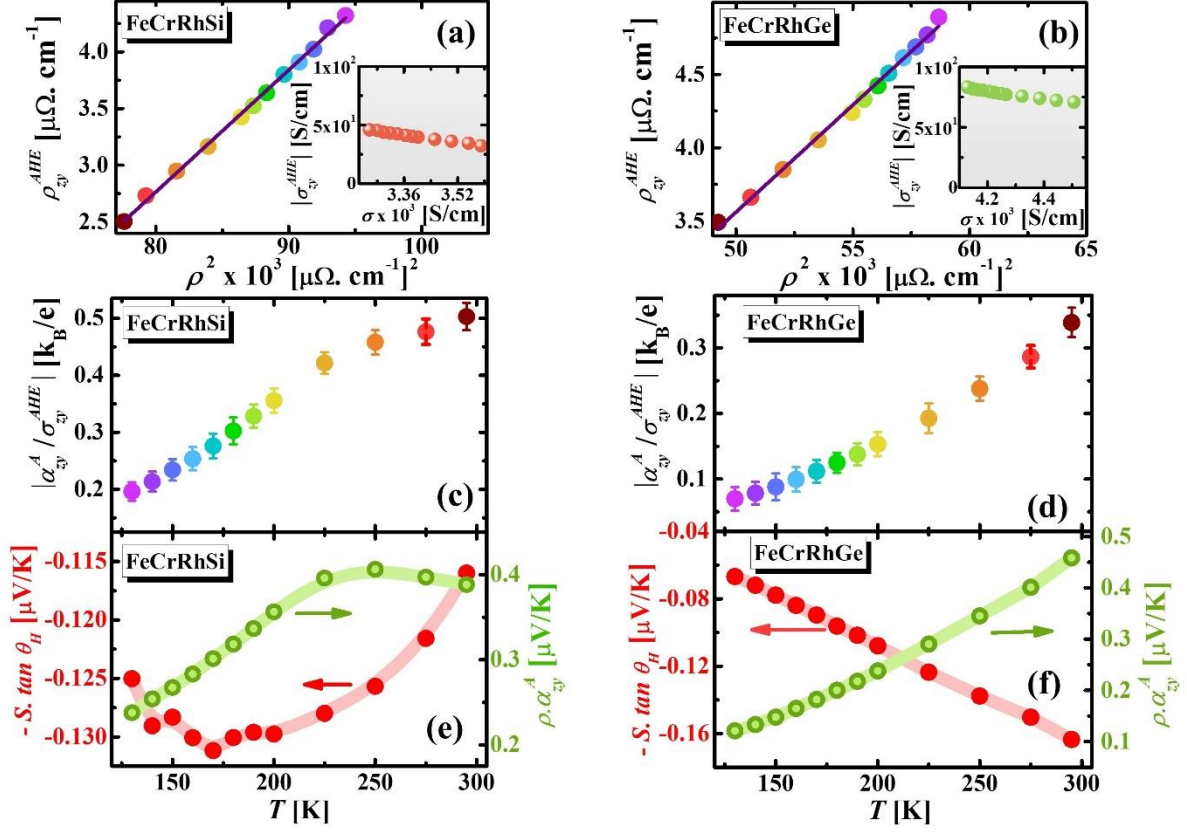


Figure 9(a) and (b): Main panels show linear fits to the ρ_{zy}^{AHE} vs. ρ^2 plots for FeCrRhSi and FeCrRhGe, respectively and insets show the corresponding plots of $|\sigma_{zy}^{AHE}|$ vs. σ for these samples. Temperature dependence of the ratio: $|\alpha_{zy}^A / \sigma_{zy}^{AHE}|$ for the samples (c) FeCrRhSi and (d) FeCrRhGe. (e) The right vertical scale displays $S_1^{ANE}(T) = \rho \cdot \alpha_{zy}^A$ and, and the left vertical scale displays $S_2^{ANE} = -S \cdot \tan \theta_H$ for FeCrRhSi. (f) The right vertical scale shows $S_1^{ANE}(T)$ and the left vertical scale shows $S_2^{ANE}(T)$ for FeCrRhGe.

Next, to confirm that both ANE and AHE in these samples share a common physical mechanism and to extract the values of intrinsic anomalous Hall conductivity $\sigma_{zy, int}^{AHE}$, we have fitted the $|\rho_{zy}^{AHE}|$ vs. ρ^2 plots for both these samples using the expression, [95,96] $\rho_{zy}^{AHE} = g(\rho_0) + (\sigma_{zy, int}^{AHE}) \cdot \rho^2$, as indicated in the main panels of **Figs. 9**(a) and (b). Here, the temperature independent term: $g(\rho_0)$ is a function of residual resistivity (ρ_0) and represents the extrinsic contributions due to skew scattering and side-jump mechanisms whereas the

temperature dependent term $(\sigma_{zy,int}^{AHE}) \cdot \rho^2$ represents the intrinsic contribution arising solely from the Berry curvature. [95,96] From the linear fits, we obtained $g(\rho_0) = -5.84 \times 10^{-6} \Omega \cdot \text{cm}$ ($-3.73 \times 10^{-6} \Omega \cdot \text{cm}$) and $|\sigma_{zy,int}^{AHE}| = 107.8 \text{ S/cm}$ (145.2 S/cm) for FeCrRhSi (FeCrRhGe). These results indicate that the Berry curvature plays the dominant role towards AHE in both these samples, whereas the negative value of $g(\rho_0)$ suggests that the extrinsic contribution is opposite to the intrinsic Karplus-Luttinger term $|\sigma_{zy,int}^{AHE}|$. [44,45,96] Notably, it is convenient to use the plot of $|\sigma_{zy}^{AHE}|$ vs. σ to empirically identify the underlying physical mechanisms responsible for anomalous Hall transport in different materials depending on the values of their σ . [96] As shown in the insets of **Figs. 9(a)** and **(b)**, the values of σ for the spin semi-metallic EQHAs FeCrRhSi and FeCrRhGe in the temperature range: $130 \text{ K} \leq T \leq 295 \text{ K}$ lie within the ranges: $3.25 \times 10^3 \text{ S/cm} \leq |\sigma| \leq 3.6 \times 10^3 \text{ S/cm}$ and $4.1 \times 10^3 \text{ S/cm} \leq |\sigma| \leq 4.6 \times 10^3 \text{ S/cm}$, respectively. These values are very much within the regime associated with the Berry curvature mechanism, [44,45,96] further confirming the intrinsic origin of both ANE and AHE in these two spin semi-metallic EQHAs.

Now, to determine the correlation between the anomalous transverse thermoelectric conduction and anomalous Hall conduction, we plot the temperature dependence of the ratio: $|\alpha_{zy}^A/\sigma_{zy}^{AHE}|$ for FeCrRhSi and FeCrRhGe, respectively in **Figs. 9(c)** and **(d)**, respectively. It is important to note that σ_{zy}^{AHE} represents the average of the Berry curvature over the occupied fermionic states. [97] In other words, σ_{zy}^{AHE} determines the transport of charge carriers and can be expressed as, $\sigma_{zy}^{AHE} \approx \frac{2\pi e^2}{ch} \langle \frac{\Omega_B}{\lambda_F^2} \rangle$; where $\langle \frac{\Omega_B}{\lambda_F^2} \rangle$ denotes the summation of the Berry curvature $\Omega_B(k)$ averaged over the cross section of the Fermi surface, λ_F is the Fermi wavelength associated with the plane perpendicular to the direction of the applied magnetic field, h is the Planck's constant, and c is the lattice parameter along the direction of applied magnetic

field. [14,97] On the other hand, α_{zy}^A represents the Berry curvature averaged over the states having a finite entropy in the vicinity of the Fermi level, i.e., within a thermal thickness of E_F . [97] Hence, α_{zy}^A represents the transport of entropy and, can be expressed as, $\alpha_{zy}^A \approx \frac{2\pi e k_B}{ch} \langle \frac{\Omega_B}{\Lambda^2} \rangle$; where, $\Lambda = \sqrt{\frac{h^2}{2\pi m_e^* k_B T}}$ is the thermal de Broglie wavelength associated with the plane perpendicular to the direction of the magnetic field. [14,97] Therefore, the ratio $|\alpha_{zy}^A / \sigma_{zy}^{AHE}|$ is governed by the quantity, $\left(\frac{k_B}{e}\right) \langle \frac{\lambda_F^2}{\Lambda^2} \rangle$. [14] Since at high temperatures, the values of λ_F and Λ become comparable in magnitude, $|\alpha_{zy}^A / \sigma_{zy}^{AHE}|$ approaches $\left(\frac{k_B}{e}\right)$ with increasing temperature [28,97]. Within the framework of Berry curvature, the value of the ratio $|\alpha_{zy}^A / \sigma_{zy}^{AHE}|$ lies between $\frac{1}{5} \left(\frac{k_B}{e}\right)$ and $\left(\frac{k_B}{e}\right)$ at room temperature in most of the topological magnets. [97] Clearly, $|\alpha_{zy}^A / \sigma_{zy}^{AHE}|$ for both the samples increases with increasing temperature and reaches $\approx (0.51 \pm 0.1) \frac{k_B}{e}$ and $\approx (0.34 \pm 0.1) \frac{k_B}{e}$ at $T = 295$ K for the spin semi-metallic EQHAs FeCrRhSi and FeCrRhGe, respectively, which are lower than that of UCo_{0.8}Ru_{0.2}Al $\left(|\alpha_{zy}^A / \sigma_{zy}^{AHE}| \approx 2 \frac{k_B}{e} \text{ at } 47 \text{ K}\right)$ [28], but closer to that for Co₂MnGa, Mn₃Sn, Co₃Sn₂S₂, CoFeVSb, all of which show Berry curvature driven anomalous Nernst thermoelectricity [20,97]. In other words, the value of the ratio $|\alpha_{zy}^A / \sigma_{zy}^{AHE}|$ for both the samples FeCrRhSi and FeCrRhGe lies between $\frac{1}{5} \left(\frac{k_B}{e}\right)$ and $\left(\frac{k_B}{e}\right)$ at room temperature, which further supports the intrinsic origin of the observed anomalous Nernst thermoelectricity in these samples within the measured temperature range. [97]

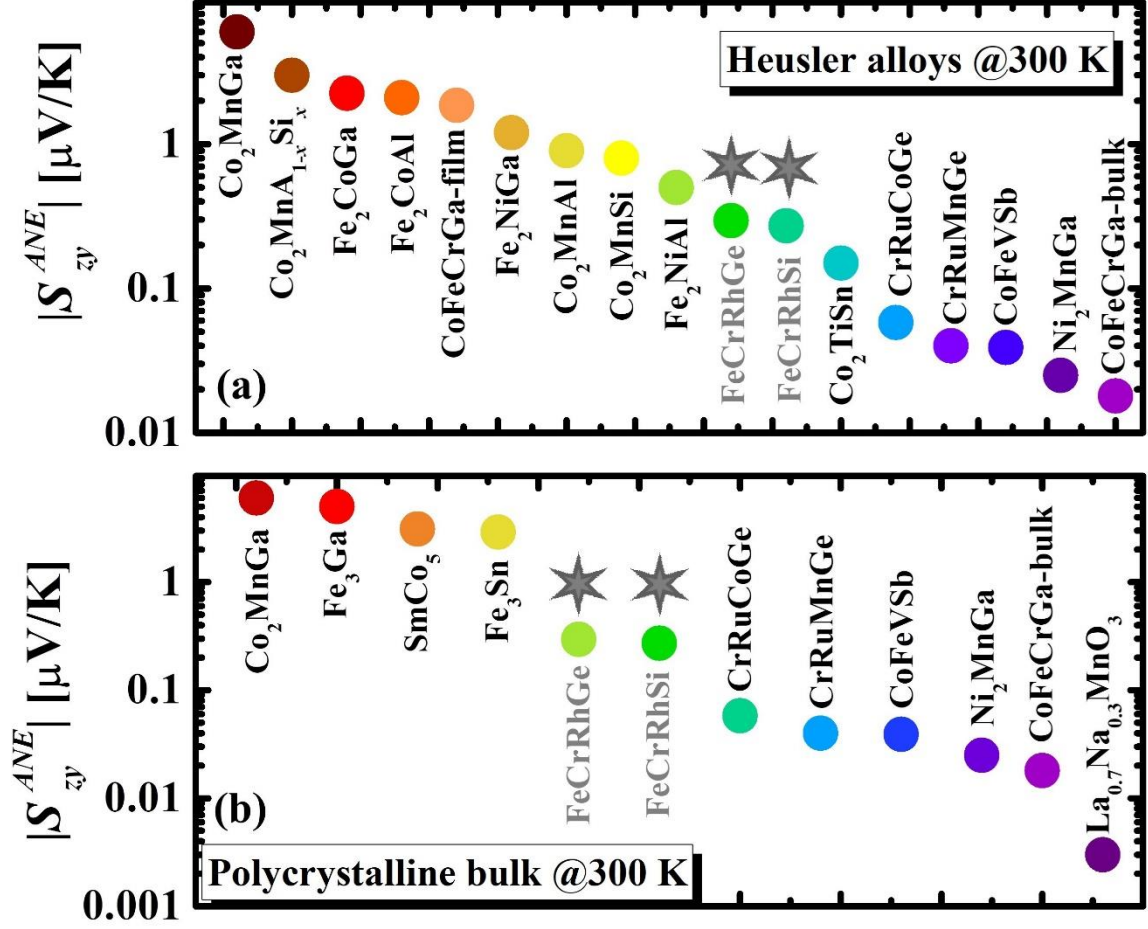


Figure 10. Comparison of the values of S_{zy}^{ANE} at room temperature for FeCrRhSi and FeCrRhGe with those of (a) different Heusler alloys and (b) different polycrystalline bulk samples reported in the literature. The samples with grey asterisks correspond to the FeCrRhX (X=Si and Ge) alloys presented in this study.

Now, with the knowledge about the temperature profiles of all the fundamental parameters, *e.g.*, $\rho(T)$, $S(T)$, $\alpha_{zy}^A(T)$, $\rho_{zy}^{AHE}(T)$, and $\sigma_{zy}^{AHE}(T)$, we can finally estimate the temperature evolution of the individual components of $S_{zy}^{ANE} = [\rho \cdot \alpha_{zy}^A - S \cdot \tan \theta_H]$, *i.e.*, $S_1^{ANE} = \rho \cdot \alpha_{zy}^A$ and, $S_2^{ANE} = -S \cdot \tan \theta_H$. [93,98] The right vertical scales of **Figs. 9(e)** and **(f)** display $S_1^{ANE}(T) = \rho \cdot \alpha_{zy}^A$ for the samples FeCrRhSi and FeCrRhGe, respectively whereas the left vertical scales of **Figs. 9(e)** and **(f)** display $S_2^{ANE}(T) = -S \cdot \tan \theta_H$ for the samples

FeCrRhSi and FeCrRhGe, respectively. While $S_1^{ANE}(T)$ for both the samples shows positive sign, $S_2^{ANE}(T)$ exhibits negative sign for both of them in the measured temperature range: $130\text{K} \leq T \leq 295\text{K}$. Moreover, $|S_1^{ANE}(T)| > |S_2^{ANE}(T)|$ for both the samples which highlights the dominating contribution of the transverse thermoelectric conduction towards the experimentally observed anomalous Nernst thermoelectricity for both FeCrRhSi and FeCrRhGe.

Next, we compare the values of S_{zy}^{ANE} for FeCrRhSi and FeCrRhGe with those of other Heusler alloys reported in the literature as shown in **Fig. 10(a)**. Evidently, the values of S_{zy}^{ANE} for both FeCrRhSi and FeCrRhGe are lower than those for single crystalline bulk [13] and thin film [99,100] samples of the semi-metallic full Heusler alloy Co_2MnGa , epitaxial thin films of Co_2MnAl [31] and Co_2MnSi [32], $\text{Co}_2\text{MnAl}_{1-x}\text{Si}_x$ thin films, [33] single crystals of Fe_2CoX ($X = \text{Al}$ and Ga), Fe_2NiX ($X = \text{Al}$ and Ga), [37] thin films of the spin gapless semiconductor CoFeCrGa , [48] *etc.*, but higher than those for the epitaxial thin films of the full Heusler alloy Co_2TiSn , [35] polycrystalline bulk sample of Ni_2MnGa shape memory alloy, [36] and other bulk polycrystalline samples of EQHAs *e.g.*, CrRuXGe ($X = \text{Co}$ and Mn), [26] CoFeVSb [20] and $\text{Co}_{1+x}\text{Fe}_{1-x}\text{CrGa}$, [12] *etc.* It is worthwhile to highlight that both FeCrRhSi and FeCrRhGe exhibit the highest values of S_{zy}^{ANE} among the EQHAs reported in the literature. Moreover, we have compared the values of S_{zy}^{ANE} for FeCrRhSi and FeCrRhGe with those of other bulk polycrystalline samples, as shown in **Fig. 10(b)**. Clearly, S_{zy}^{ANE} for both FeCrRhSi and FeCrRhGe are lower than those for bulk polycrystalline full Heusler alloy Co_2MnGa , [101] polycrystalline topological ferromagnets Fe_3Ga , [102] permanent magnet SmCo_5 , [103] polycrystalline iron-based Kagome ferromagnet Fe_3Sn , [15] but much higher than those of other bulk polycrystalline samples of the EQHA family, [12,20,26] *e.g.*, polycrystalline bulk sample of Ni_2MnGa , [36] half metallic ferromagnetic hole-doped manganite

$\text{La}_{0.7}\text{Na}_{0.3}\text{MnO}_3$, [22] *etc.* Therefore, both the EQHAs FeCrRhSi and FeCrRhGe hold great promises towards the development of room temperature ANE based thermopile device applications.

4. Conclusions

To summarize, we have performed a comprehensive investigation of magnetic, electrical, thermal and transverse magneto-thermoelectric properties of the topological spin semi-metallic quaternary Heusler alloys FeCrRhSi and FeCrRhGe. Magnetic measurements revealed room temperature ferromagnetic behavior with high Curie temperatures ($T_c > 400$ K) of these samples wherein energetically competing FM and AFM phases coexist at low temperatures, giving rise to field-induced metamagnetic transitions. The electrical resistivity of both samples exhibited semi-metallic characteristics throughout the measured temperature range, along with the low temperature resistivity upturns driven by Kondo-like scattering. The negative sign of the longitudinal Seebeck coefficient throughout the measured temperature range in both FeCrRhSi and FeCrRhGe indicates that electrons are the majority carriers for the thermally driven charge transport in these samples. Most importantly, both samples exhibited giant values of S_{zy}^{ANE} at room temperature: S_{zy}^{ANE} for FeCrRhSi and FeCrRhGe at 295 K are $(0.272 \pm 0.02) \mu\text{V} \cdot \text{K}^{-1}$ and $(0.295 \pm 0.02) \mu\text{V} \cdot \text{K}^{-1}$, respectively, both of which are nearly three times higher than those of the best EQHA reported so far. Our in-depth analysis has revealed that the Berry curvature governs such large ANE in these topological spin semi-metallic EQHAs. We have further highlighted that the contribution of the transverse thermoelectric conduction to the observed large ANE in both samples dominates over that of the anomalous Hall effect affecting the longitudinal Seebeck effect driven charge carrier flow. The ratio, $|\alpha_{zy}^A/\sigma_{zy}^{AHE}|$ for both these samples lies between $0.2 \left(\frac{k_B}{e}\right)$ and $\left(\frac{k_B}{e}\right)$ at room temperature, further confirming intrinsic Berry curvature as the underlying physical origin of the observed large ANEs.

5. Acknowledgements

Financial support by the US Department of Energy, Office of Basic Energy Sciences, Division of Materials Science and Engineering under Award No. DE-FG02-07ER46438. HS also thanks support from IIT Bombay for a short-term visiting professorship. KGS thanks DST, Govt. of India for funding via Indo-Russian project DST/INT/RUS/RSF/P-47/2021. NS would like to acknowledge support from the US Department of Defense SMART Scholarship program.

References

- [1] G. E. W. Bauer, E. Saitoh, and B. J. Van Wees, *Spin Caloritronics*, Nat. Mater. **11**, 391 (2012).
- [2] Y. Pu, D. Chiba, F. Matsukura, H. Ohno, and J. Shi, *Mott Relation for Anomalous Hall and Nernst Effects in Ga 1- x Mn x As Ferromagnetic Semiconductors*, Phys. Rev. Lett. **101**, 117208 (2008).
- [3] K. Uchida, W. Zhou, and Y. Sakuraba, *Transverse Thermoelectric Generation Using Magnetic Materials*, Appl. Phys. Lett. **118**, 140504 (2021).
- [4] Y. Pan, C. Le, B. He, S. J. Watzman, M. Yao, J. Gooth, J. P. Heremans, Y. Sun, and C. Felser, *Giant Anomalous Nernst Signal in the Antiferromagnet YbMnBi₂*, Nat. Mater. **21**, 203 (2022).
- [5] S. Noguchi, K. Fujiwara, Y. Yanagi, M.-T. Suzuki, T. Hirai, T. Seki, K. Uchida, and A. Tsukazaki, *Bipolarity of Large Anomalous Nernst Effect in Weyl Magnet-Based Alloy Films*, Nat. Phys. **1** (2024).
- [6] N. Nagaosa, J. Sinova, S. Onoda, A. H. MacDonald, and N. P. Ong, *Anomalous Hall Effect*, Rev. Mod. Phys. **82**, 1539 (2010).
- [7] S. N. Guin et al., *Anomalous Nernst Effect beyond the Magnetization Scaling Relation in the Ferromagnetic Heusler Compound Co₂MnGa*, NPG Asia Mater. **11**, 1 (2019).
- [8] H. Yang et al., *Giant Anomalous Nernst Effect in the Magnetic Weyl Semimetal Co₃Sn₂S₂*, Phys. Rev. Mater. **4**, 24202 (2020).
- [9] J. Xu, W. A. Phelan, and C.-L. Chien, *Large Anomalous Nernst Effect in a van Der Waals Ferromagnet Fe₃GeTe₂*, Nano Lett. **19**, 8250 (2019).
- [10] T. Yamazaki, T. Seki, R. Modak, K. Nakagawara, T. Hirai, K. Ito, K. Uchida, and K. Takanashi, *Thickness Dependence of Anomalous Hall and Nernst Effects in Ni-Fe Thin Films*, Phys. Rev. B **105**, 214416 (2022).

- [11] H. Nakayama, K. Masuda, J. Wang, A. Miura, K. Uchida, M. Murata, and Y. Sakuraba, *Mechanism of Strong Enhancement of Anomalous Nernst Effect in Fe by Ga Substitution*, Phys. Rev. Mater. **3**, 114412 (2019).
- [12] A. Chanda, D. Rani, J. Nag, A. Alam, K. G. Suresh, M. H. Phan, and H. Srikanth, *Emergence of Asymmetric Skew-Scattering Dominated Anomalous Nernst Effect in the Spin Gapless Semiconductors $\text{Co}_{1+x}\text{Fe}_{1-x}\text{CrGa}$* , Phys. Rev. B **106**, 134416 (2022).
- [13] A. Sakai et al., *Giant Anomalous Nernst Effect and Quantum-Critical Scaling in a Ferromagnetic Semimetal*, Nat. Phys. **14**, 1119 (2018).
- [14] L. Ding et al., *Intrinsic Anomalous Nernst Effect Amplified by Disorder in a Half-Metallic Semimetal*, Phys. Rev. X **9**, 41061 (2019).
- [15] T. Chen et al., *Large Anomalous Nernst Effect and Nodal Plane in an Iron-Based Kagome Ferromagnet*, Sci. Adv. **8**, eabk1480 (2022).
- [16] D. Khadka, T. R. Thapaliya, S. H. Parra, J. Wen, R. Need, J. M. Kikkawa, and S. X. Huang, *Anomalous Hall and Nernst Effects in Epitaxial Films of Topological Kagome Magnet Fe_3Sn_2* , Phys. Rev. Mater. **4**, 84203 (2020).
- [17] A. Sakai et al., *Iron-Based Binary Ferromagnets for Transverse Thermoelectric Conversion*, Nature **581**, 53 (2020).
- [18] M. Li et al., *Large Anomalous Nernst Effects at Room Temperature in Fe_3Pt Thin Films*, Adv. Mater. **35**, 2301339 (2023).
- [19] M. Ikhlas, T. Tomita, T. Koretsune, M.-T. Suzuki, D. Nishio-Hamane, R. Arita, Y. Otani, and S. Nakatsuji, *Large Anomalous Nernst Effect at Room Temperature in a Chiral Antiferromagnet*, Nat. Phys. **13**, 1085 (2017).
- [20] A. Chanda, J. Nag, A. Alam, K. G. Suresh, M.-H. Phan, and H. Srikanth, *Intrinsic Berry Curvature Driven Anomalous Nernst Thermopower in the Semimetallic Heusler*

- Alloy CoFeVSb*, Phys. Rev. B **107**, L220403 (2023).
- [21] C. T. Bui and F. Rivadulla, *Anomalous and Planar Nernst Effects in Thin Films of the Half-Metallic Ferromagnet $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$* , Phys. Rev. B **90**, 100403 (2014).
- [22] A. Ghosh, R. Das, and R. Mahendiran, *Skew Scattering Dominated Anomalous Nernst Effect in $\text{La}_{1-x}\text{Na}_x\text{MnO}_3$* , J. Appl. Phys. **125**, 153902 (2019).
- [23] A. Ghosh, A. Chanda, and R. Mahendiran, *Anomalous Nernst Effect in $\text{Pr}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$* , AIP Adv. **11**, 35031 (2021).
- [24] A. Ghosh, A. Chanda, M. Manikandan, and R. Mahendiran, *Rare Earth Size Dependence of Nernst Thermopower in Ferromagnetic Perovskites: $\text{R}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$ ($\text{R} = \text{La, Pr, and Nd}$)*, J. Magn. Magn. Mater. **537**, 168240 (2021).
- [25] T. Miyasato, N. Abe, T. Fujii, A. Asamitsu, S. Onoda, Y. Onose, N. Nagaosa, and Y. Tokura, *Crossover Behavior of the Anomalous Hall Effect and Anomalous Nernst Effect in Itinerant Ferromagnets*, Phys. Rev. Lett. **99**, 86602 (2007).
- [26] A. Chanda, J. Nag, N. Schulz, A. Alam, K. G. Suresh, M.-H. Phan, and H. Srikanth, *Large Anomalous Nernst Effect and Its Bipolarity in the Quaternary Equiatomic Heusler Alloys CrRu_XGe ($X = \text{Co and Mn}$)*, Phys. Rev. B **109**, 224415 (2024).
- [27] R. Ramos, M. H. Aguirre, A. Anadón, J. Blasco, I. Lucas, K. Uchida, P. A. Algarabel, L. Morellón, E. Saitoh, and M. R. Ibarra, *Anomalous Nernst Effect of Fe_3O_4 Single Crystal*, Phys. Rev. B **90**, 54422 (2014).
- [28] T. Asaba, V. Ivanov, S. M. Thomas, S. Y. Savrasov, J. D. Thompson, E. D. Bauer, and F. Ronning, *Colossal Anomalous Nernst Effect in a Correlated Noncentrosymmetric Kagome Ferromagnet*, Sci. Adv. **7**, eabf1467 (2021).
- [29] A. Hirohata and D. C. Lloyd, *Heusler Alloys for Metal Spintronics*, MRS Bull. **47**, 593 (2022).
- [30] S. Tavares, K. Yang, and M. A. Meyers, *Heusler Alloys: Past, Properties, New Alloys*,

- and Prospects*, Prog. Mater. Sci. **132**, 101017 (2023).
- [31] A. T. Breidenbach, H. Yu, T. A. Peterson, A. P. McFadden, W. K. Peria, C. J. Palmstrøm, and P. A. Crowell, *Anomalous Nernst and Seebeck Coefficients in Epitaxial Thin Film $\text{Co}_2\text{MnAl}_x\text{Si}_{1-x}$ and Co_2FeAl* , Phys. Rev. B **105**, 144405 (2022).
- [32] C. D. W. Cox, A. J. Caruana, M. D. Cropper, and K. Morrison, *Anomalous Nernst Effect in Co_2MnSi Thin Films*, J. Phys. D: Appl. Phys. **53**, 35005 (2019).
- [33] Y. Sakuraba, K. Hyodo, A. Sakuma, and S. Mitani, *Giant Anomalous Nernst Effect in the $\text{Co}_2\text{MnAl}_{1-x}\text{Si}_x$ Heusler Alloy Induced by Fermi Level Tuning and Atomic Ordering*, Phys. Rev. B **101**, 134407 (2020).
- [34] J. Hu et al., *Regulating the Anomalous Hall and Nernst Effects in Heusler-Based Trilayers*, Appl. Phys. Lett. **117**, 62405 (2020).
- [35] J. Hu et al., *Anomalous Hall and Nernst Effects in Co_2TiSn and $\text{Co}_2\text{Ti}_{0.6}\text{V}_{0.4}\text{Sn}$ Heusler Thin Films*, Phys. Rev. Appl. **10**, 44037 (2018).
- [36] A. De, A. K. Singh, S. Singh, and S. Nair, *Temperature Dependence of the Anomalous Nernst Effect in Ni_2MnGa Shape Memory Alloy*, Phys. Rev. B **103**, L020404 (2021).
- [37] F. Mende, J. Noky, S. N. Guin, G. H. Fecher, K. Manna, P. Adler, W. Schnelle, Y. Sun, C. Fu, and C. Felser, *Large Anomalous Hall and Nernst Effects in High Curie-Temperature Iron-Based Heusler Compounds*, Adv. Sci. **8**, 2100782 (2021).
- [38] S. A. Khandy and J.-D. Chai, *Thermoelectric Properties, Phonon, and Mechanical Stability of New Half-Metallic Quaternary Heusler Alloys: FeRhCrZ ($Z = \text{Si}$ and Ge)*, J. Appl. Phys. **127**, (2020).
- [39] V. Alijani et al., *Electronic, Structural, and Magnetic Properties of the Half-Metallic Ferromagnetic Quaternary Heusler Compounds CoFeMnZ ($Z = \text{Al}, \text{Ga}, \text{Si}, \text{Ge}$)*, Phys. Rev. B **84**, 224416 (2011).

- [40] D. Rani, L. Bainsla, K. G. Suresh, A. Alam, and others, *Spin-Gapless Semiconducting Nature of Co-Rich $\text{Co}_{1+x}\text{Fe}_{1-x}\text{CrGa}$* , Phys. Rev. B **99**, 104429 (2019).
- [41] L. Bainsla, A. I. Mallick, M. M. Raja, A. K. Nigam, B. S. D. C. S. Varaprasad, Y. K. Takahashi, A. Alam, K. G. Suresh, and K. Hono, *Spin Gapless Semiconducting Behavior in Equiatomic Quaternary CoFeMnSi Heusler Alloy*, Phys. Rev. B **91**, 104408 (2015).
- [42] S. Ouardi, G. H. Fecher, C. Felser, and J. Kübler, *Realization of Spin Gapless Semiconductors: The Heusler Compound Mn_2CoAl* , Phys. Rev. Lett. **110**, 100401 (2013).
- [43] J. Nag et al., *CoFeVSb : A Promising Candidate for Spin Valve and Thermoelectric Applications*, Phys. Rev. B **105**, 144409 (2022).
- [44] Y. Venkateswara, S. S. Samatham, P. D. Babu, K. G. Suresh, and A. Alam, *Coexistence of Spin Semimetal and Weyl Semimetal Behavior in FeRhCrGe* , Phys. Rev. B **100**, 180404 (2019).
- [45] Y. Venkateswara, J. Nag, S. S. Samatham, A. K. Patel, P. D. Babu, M. R. Varma, J. Nayak, K. G. Suresh, and A. Alam, *FeRhCrSi : Spin Semimetal with Spin-Valve Behavior at Room Temperature*, Phys. Rev. B **107**, L100401 (2023).
- [46] J. Nag, R. Venkatesh, A. Jha, P. Stamenov, P. D. Babu, A. Alam, and S. Gopinatha Warrier, *Topological Spin Semimetal Feature in a Fully Compensated Ferrimagnet CoRuVSi* , ACS Appl. Electron. Mater. **5**, 5944 (2023).
- [47] J. Nag, D. Rani, J. Kangsabanik, D. Singh, R. Venkatesh, P. D. Babu, K. G. Suresh, and A. Alam, *Bipolar Magnetic Semiconducting Behavior in VNbRuAl* , Phys. Rev. B **104**, 134406 (2021).
- [48] A. Chanda, D. Rani, D. DeTellem, N. Alzahrani, D. A. Arena, S. Witanachchi, R. Chatterjee, M.-H. Phan, and H. Srikanth, *Large Thermo-Spin Effects in Heusler Alloy-*

- Based Spin Gapless Semiconductor Thin Films*, ACS Appl. Mater. Interfaces **15**, 53697 (2023).
- [49] A. Chanda, C. Holzmann, N. Schulz, J. Seyd, M. Albrecht, M.-H. Phan, and H. Srikanth, *Scaling of the Thermally Induced Sign Inversion of Longitudinal Spin Seebeck Effect in a Compensated Ferrimagnet: Role of Magnetic Anisotropy*, Adv. Funct. Mater. **32**, 2109170 (2022).
- [50] K. He, N. Chen, C. Wang, L. Wei, and J. Chen, *Method for Determining Crystal Grain Size by X-Ray Diffraction*, Cryst. Res. Technol. **53**, 1700157 (2018).
- [51] B. Shi, J. Li, C. Jin, J. Yang, C. Zhang, Y. Yan, Y. Wang, and G. Zhang, *Distinct Transport Behaviors and Electronic Structures in Heusler Alloys CoFeCrGa and CoFeCrAl*, J. Magn. Magn. Mater. **517**, 167383 (2021).
- [52] L. Bainsla, A. I. Mallick, M. M. Raja, A. A. Coelho, A. K. Nigam, D. D. Johnson, A. Alam, and K. G. Suresh, *Origin of Spin Gapless Semiconductor Behavior in CoFeCrGa: Theory and Experiment*, Phys. Rev. B **92**, 45201 (2015).
- [53] U. Köbler, A. Hoser, and W. Schäfer, *On the Temperature Dependence of the Magnetic Excitations*, Phys. B Condens. Matter **364**, 55 (2005).
- [54] U. Krey, *On the Significance of Quantum Effects and Interactions for the Apparent Universality of Bloch Laws for $M_s(T)$* , J. Magn. Magn. Mater. **268**, 277 (2004).
- [55] U. Köbler, *Temperature Dependence of the Spontaneous Magnetization of Body-Centred-Cubic Bulk Iron, Amorphous Iron and Thin Iron Films*, J. Phys. Condens. Matter **14**, 8861 (2002).
- [56] R. Peña-Garcia, Y. Guerra, F. R. de Souza, L. A. P. Gonçalves, and E. Padrón-Hernández, *The Extended Bloch's Law in Yttrium Iron Garnet Doped with Zn, Ni and Co*, Phys. E Low-Dimensional Syst. Nanostructures **103**, 354 (2018).
- [57] I. Galanakis, P. H. Dederichs, and N. Papanikolaou, *Slater-Pauling Behavior and*

- Origin of the Half-Metallicity of the Full-Heusler Alloys*, Phys. Rev. B **66**, 174429 (2002).
- [58] A. Chanda, J. E. Shoup, N. Schulz, D. A. Arena, and H. Srikanth, *Tunable Competing Magnetic Anisotropies and Spin Reconfigurations in Ferrimagnetic Fe_{100-x}Gd_x Alloy Films*, Phys. Rev. B **104**, 94404 (2021).
- [59] V. K. Sharma, M. K. Chattopadhyay, and S. B. Roy, *Kinetic Arrest of the First Order Austenite to Martensite Phase Transition in Ni₅₀Mn₃₄In₁₆: Dc Magnetization Studies*, Phys. Rev. B **76**, 140401 (2007).
- [60] Ö. Çakır, M. Acet, M. Farle, E. Dias, and K. Priolkar, *Kinetic Arrest in Magnetically Inhomogeneous C-Deficient Mn₃GaC*, J. Magn. Magn. Mater. **390**, 96 (2015).
- [61] M. Vasundhara, V. Srinivas, and V. V Rao, *Electronic Transport in Heusler-Type Fe₂VA1_{1-x}M_x Alloys (M= B, In, Si)*, Phys. Rev. B—Condensed Matter Mater. Phys. **77**, 224415 (2008).
- [62] N. Reumann, A. Riss, F. Garmroudi, M. Parzer, J. Kovacevic, T. Mori, and E. Bauer, *Thermoelectric Properties and Low-Temperature Transport Anomalies in the p-Type Full-Heusler Compounds Fe_{2-x}Cr_xVA1*, Phys. Rev. B **106**, 235138 (2022).
- [63] J. Kondo, *Resistance Minimum in Dilute Magnetic Alloys*, Prog. Theor. Phys. **32**, 37 (1964).
- [64] D. S. Rana, J. H. Markna, R. N. Parmar, D. G. Kuberkar, P. Raychaudhuri, J. John, and S. K. Malik, *Low-Temperature Transport Anomaly in the Magnetoresistive Compound (La_{0.5}Pr_{0.2})Ba_{0.3}MnO₃*, Phys. Rev. B **71**, 212404 (2005).
- [65] S. Chatterjee, S. Chatterjee, S. Giri, and S. Majumdar, *Transport Properties of Heusler Compounds and Alloys*, J. Phys. Condens. Matter (2021).
- [66] K. Yamada, M. Nakano, K. Yoshida, K. Hanzawa, and A. Sakurai, *T-Square Term of Electrical Resistivity*, Prog. Theor. Phys. **82**, 689 (1989).

- [67] A. Urushibara, Y. Moritomo, T. Arima, A. Asamitsu, G. Kido, and Y. Tokura, *Insulator-Metal Transition and Giant Magnetoresistance in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$* , Phys. Rev. B **51**, 14103 (1995).
- [68] K. Kubo and N. Ohata, *A Quantum Theory of Double Exchange. I*, J. Phys. Soc. Japan **33**, 21 (1972).
- [69] J. M. Ziman, *Principles of the Theory of Solids* (Cambridge university press, 1972).
- [70] D. Bombor, C. G. F. Blum, O. Volkonskiy, S. Rodan, S. Wurmehl, C. Hess, and B. Büchner, *Half-Metallic Ferromagnetism with Unexpectedly Small Spin Splitting in the Heusler Compound Co_2FeSi* , Phys. Rev. Lett. **110**, 66601 (2013).
- [71] J. Zhang, Y. Xu, S. Cao, G. Cao, Y. Zhang, and C. Jing, *Kondo-like Transport and Its Correlation with the Spin-Glass Phase in Perovskite Manganites*, Phys. Rev. B **72**, 54410 (2005).
- [72] A. Ślebarski, M. Fijałkowski, J. Deniszczuk, M. M. Maśka, and D. Kaczorowski, *Off-Stoichiometric Effect on Magnetic and Electron Transport Properties of $\text{Fe}_{2-\text{VAl}}$ 1.35 and $\text{Ni}_{2-\text{VAl}}$: A Comparative Study*, Phys. Rev. B **109**, 165105 (2024).
- [73] B. Xu and M. J. Verstraete, *First Principles Explanation of the Positive Seebeck Coefficient of Lithium*, Phys. Rev. Lett. **112**, 196603 (2014).
- [74] F. J. DiSalvo, *Thermoelectric Cooling and Power Generation*, Science (80-.). **285**, 703 (1999).
- [75] J. L. Cohn, J. J. Neumeier, C. P. Popoviciu, K. J. McClellan, and T. Leventouri, *Local Lattice Distortions and Thermal Transport in Perovskite Manganites*, Phys. Rev. B **56**, R8495 (1997).
- [76] T. M. Tritt, *Thermoelectric Phenomena, Materials, and Applications*, Annu. Rev. Mater. Res. **41**, 433 (2011).
- [77] H.-S. Kim, Z. M. Gibbs, Y. Tang, H. Wang, and G. J. Snyder, *Characterization of*

- Lorenz Number with Seebeck Coefficient Measurement*, APL Mater. **3**, 41506 (2015).
- [78] J. Callaway, *Model for Lattice Thermal Conductivity at Low Temperatures*, Phys. Rev. **113**, 1046 (1959).
- [79] J. Callaway, *Low-Temperature Lattice Thermal Conductivity*, Phys. Rev. **122**, 787 (1961).
- [80] S. Bhattacharya, A. Mehdizadeh Dehkordi, S. Tennakoon, R. Adebisi, J. R. Gladden, T. Darroudi, H. N. Alshareef, and T. M. Tritt, *Role of Phonon Scattering by Elastic Strain Field in Thermoelectric Sr_{1-x}Y_xTiO₃*, J. Appl. Phys. **115**, 223712 (2014).
- [81] P. G. Klemens, *Thermal Resistance Due to Point Defects at High Temperatures*, Phys. Rev. **119**, 507 (1960).
- [82] A. Petersen, S. Bhattacharya, T. M. Tritt, and S. J. Poon, *Critical Analysis of Lattice Thermal Conductivity of Half-Heusler Alloys Using Variations of Callaway Model*, J. Appl. Phys. **117**, (2015).
- [83] G. A. Slack and S. Galginaitis, *Thermal Conductivity and Phonon Scattering by Magnetic Impurities in CdTe*, Phys. Rev. **133**, A253 (1964).
- [84] C. J. Glassbrenner and G. A. Slack, *Thermal Conductivity of Silicon and Germanium from 3 K to the Melting Point*, Phys. Rev. **134**, A1058 (1964).
- [85] J. Yang, D. T. Morelli, G. P. Meisner, W. Chen, J. S. Dyck, and C. Uher, *Influence of Electron-Phonon Interaction on the Lattice Thermal Conductivity of Co_{1-x}Ni_xSb₃*, Phys. Rev. B **65**, 94115 (2002).
- [86] C. S. Lue, C. F. Chen, J. Y. Lin, Y. T. Yu, and Y. K. Kuo, *Thermoelectric Properties of Quaternary Heusler Alloys Fe₂VAl_{1-x}Si_x*, Phys. Rev. B—Condensed Matter Mater. Phys. **75**, 64204 (2007).
- [87] *Supporting Information Available Online*, (n.d.).
- [88] K. Behnia, *Fundamentals of Thermoelectricity* (OUP Oxford, 2015).

- [89] A. C. Anderson, R. B. Rauch, and M. M. Kreitman, *Another Comparison of Thermal Bonding Agents*, Rev. Sci. Instrum. **41**, 469 (1970).
- [90] E. Gmelin, M. Asen-Palmer, M. Reuther, and R. Villar, *Thermal Boundary Resistance of Mechanical Contacts between Solids at Sub-Ambient Temperatures*, J. Phys. D. Appl. Phys. **32**, R19 (1999).
- [91] S. Jeong and C. Park, *Thermal Contact Problems at Cryogenic Temperature*, Prog. Supercond. Cryog. **17**, 1 (2015).
- [92] T. Ashworth, J. E. Loomer, and M. M. Kreitman, *Thermal Conductivity of Nylons and Apiezon Greases*, in *Advances in Cryogenic Engineering* (Springer, 1973), pp. 271–279.
- [93] Y. Wang, A. Sakai, S. Minami, H. Gu, T. Chen, Z. Feng, D. Nishio-Hamane, and S. Nakatsuji, *Robust Giant Anomalous Nernst Effect in Polycrystalline Nodal Web Ferromagnets*, Appl. Phys. Lett. **125**, (2024).
- [94] W. Zhou, K. Masuda, and Y. Sakuraba, *Origin of Negative Anomalous Nernst Thermopower in Mn-Ga Ordered Alloys*, Appl. Phys. Lett. **118**, (2021).
- [95] S. Bera, S. Chatterjee, S. Pradhan, S. K. Pradhan, S. Kalimuddin, A. Bera, A. K. Nandy, and M. Mondal, *Anomalous Hall Effect Induced by Berry Curvature in the Topological Nodal-Line van Der Waals Ferromagnet Fe_4GeTe_2* , Phys. Rev. B **108**, 115122 (2023).
- [96] Y. Han, C. Qiu, W. Ren, Y. Wu, L. Jiang, X. Luo, C. Chen, C. Fang, and S. Ma, *Anomalous Hall and Nernst Effects in the Half-Metallic Ferromagnet Gd_4Sb_3* , Phys. Rev. B **110**, 144405 (2024).
- [97] L. Xu, X. Li, L. Ding, T. Chen, A. Sakai, B. Fauqué, S. Nakatsuji, Z. Zhu, and K. Behnia, *Anomalous Transverse Response of Co_2MnGa and Universality of the Room-Temperature ρ_{xy}/ρ_{xx} Ratio across Topological Magnets*, Phys. Rev. B **101**,

- 180404 (2020).
- [98] Z. Liu, G. Li, H. Fu, and Y. Lu, *Robust Room-Temperature Anomalous Nernst Effect over a Broad Thickness and Composition Range in Chemically Disordered FePt_{100-x} Films*, Appl. Phys. Lett. **124**, (2024).
 - [99] H. Reichlova et al., *Large Anomalous Nernst Effect in Thin Films of the Weyl Semimetal Co₂MnGa*, Appl. Phys. Lett. **113**, 212405 (2018).
 - [100] R. Uesugi, T. Higo, and S. Nakatsuji, *Giant Anomalous Nernst Effect in Polycrystalline Thin Films of the Weyl Ferromagnet Co₂MnGa*, Appl. Phys. Lett. **123**, (2023).
 - [101] W. Zhou, A. Miura, T. Hirai, Y. Sakuraba, and K. Uchida, *Seebeck-Driven Transverse Thermoelectric Generation in Magnetic Hybrid Bulk Materials*, Appl. Phys. Lett. **122**, (2023).
 - [102] Z. Feng, S. Minami, S. Akamatsu, A. Sakai, T. Chen, D. Nishio-Hamane, and S. Nakatsuji, *Giant and Robust Anomalous Nernst Effect in a Polycrystalline Topological Ferromagnet at Room Temperature*, Adv. Funct. Mater. **32**, 2206519 (2022).
 - [103] A. Miura, K. Masuda, T. Hirai, R. Iguchi, T. Seki, Y. Miura, H. Tsuchiura, K. Takanashi, and K. Uchida, *High-Temperature Dependence of Anomalous Ettingshausen Effect in SmCo₅-Type Permanent Magnets*, Appl. Phys. Lett. **117**, (2020).