



Substrate-dependent magneto-thermoelectric properties in FeRh thin films during antiferromagnetic–ferromagnetic phase transition

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FeRh undergoes a first-order antiferromagnetic–ferromagnetic (AFM–FM) transition near room temperature that strongly affects electronic transport. Bulk FeRh shows large but localized Thomson responses near the thermally induced transition. Here, we show that this behavior is tunable in thin films. We measure the thermomagnetic responses of near-equimolar FeRh films (20–80 nm) grown on Al₂O₃, SiO₂/Si, and MgO. For films grown on the same substrate, increasing thickness reduces strain and sharpens the AFM–FM transition, yielding a correspondingly sharper Thomson response. Thicker films reach peaks exceeding $-250 \mu\text{V K}^{-1}$ over narrow temperature ranges, whereas thinner films display smaller peaks spanning up to ~ 100 K. Changing the substrate alters crystallinity, microstructure, and strain, leading to pronounced differences in the baseline Seebeck coefficient in both phases. These results demonstrate that strain and microstructure enable engineering the magnitude and temperature span of the Thomson effect in FeRh thin films.

Introduction

Solid–solid phase-change materials (PCMs) undergo reversible transformations between distinct structural or magnetic states, accompanied by abrupt changes in physical properties that can be exploited for sensing, information storage, and energy conversion applications [1–8]. Among PCMs, magnetostructural transitions are particularly attractive because they enable strong, reversible modifications of electronic transport in response to temperature, magnetic field, or strain [9–13]. In addition to well-studied magnetoresistive and magnetocaloric responses [14–19], magnetic phase transitions can give rise to pronounced thermomagnetic transport effects, including sharp variations in the Seebeck coefficient and an enhanced Thomson response. The Thomson effect, which describes reversible heat absorption or

release in a current-carrying conductor subjected to a temperature gradient, is generally weak in conventional materials but can become substantial near first-order magnetic transitions, where transport coefficients vary rapidly with temperature. Understanding how such transition-driven Thomson responses can be controlled in thin films is therefore of both fundamental and practical interest.

The binary intermetallic alloy FeRh with a CsCl-type (B2) structure exhibits a first-order magnetostructural phase transition near room temperature [20]. Its near-equimolar compositional simplicity and transition temperature make it relevant for semiconductor industry applications such as antiferromagnetic memory [21, 22] and fast switching [23–25]. Upon heating, FeRh transitions from the antiferromagnetic (AFM, α'') phase to the

ferromagnetic (FM, α') phase with an approximately 0.3% isotropic lattice expansion, preserving its original cubic CsCl structure [26]. Many studies have been conducted to elucidate the fundamental aspects underlying this FeRh transition [27–36].

Hard X-ray photoemission has revealed distinct differences between the AFM and FM states throughout the entire valence-band spectrum and was elucidated by density functional theory (DFT) calculations [28]. Recent work proposes that the phase transition in FeRh may be driven by the competing magnetic states of the Rh atom, which determine the magnetic state of the system [29, 30]. Using first-principles calculations (non-collinear version of the linear muffin-tin orbital method), Gu and Antropov evaluated the AFM–FM free-energy difference and concluded that magnon (spin-wave) excitations primarily govern the transition [31]. Complementarily, Barker and Chantrell leveraged atomistic spin dynamics to demonstrate that the first-order FeRh phase transition can arise from the competition between bilinear and higher-order four-spin exchange terms in the effective spin Hamiltonian [32]. External control parameters have been extensively employed to tune the transition—including composition, magnetic field, thickness, and doping [26, 37–41]. For example, minimal compositional variation ($0.48 \leq x < 0.54$ for $\text{Fe}_{1-x}\text{Rh}_x$) preserves the phase transition while shifting its temperature. Introducing various dopants, such as Ni, Au, Cu, Pt, and Pd, can also modify the phase transition temperature [42–45]. Finally, applying tensile (compressive) strain has been shown to shift the transition to lower (higher) temperatures [46].

Despite the prolonged investigation of FeRh, only a handful of studies have been conducted to explore its potential in thermal-to-electrical energy conversion using the three thermoelectric phenomena: the Seebeck effect, the Thomson effect, and the Nernst effect. The Seebeck effect generates a longitudinal voltage gradient (∇V) along the direction of the applied temperature gradient (∇T) and is the basis of conventional thermoelectric devices. The Thomson effect describes heat absorption or release when current flows through a material with a ∇T . In the Nernst effect, a transverse voltage difference (ΔV) develops in response to mutually perpendicular ∇T and magnetic field. Studying the Hall coefficient, heat capacity, and Seebeck coefficient in bulk FeRh during the phase transition, Kobayashi et al. reported a significant reconstruction of the Fermi surface [47]. Modak et al. observed a giant Thomson effect in Ni-doped bulk FeRh, which can result in significant Thomson cooling that offsets the Joule heating [48]. Yamauchi et al. studied the compositional dependence of the Nernst effect in FeRh/SiO₂/Si films at room temperature and reported a maximum Nernst response when Rh is 48%, which has a ferromagnetic state close to the phase transition [49]. Using anomalous Nernst microscopy, Gray et al. [50] reported a positive Nernst response in Pt/FeRh/MgO films while using a conventional probe setup. Saglam et al. [51] observed a sign change of the Nernst coefficient (N/B) in a

FeRh/MgO film around the phase transition, where B denotes the magnetic field. A theoretical explanation of this temperature-dependent sign change in N/B in FeRh is still missing.

Despite the large literature, a substrate-resolved, thin-film map of Seebeck, Thomson, and Nernst responses has not been systematically reported, and a quantitative account of AFM-phase Nernst sign behavior in near-equi-molar FeRh films remains limited. In this work, we investigate near-equi-molar FeRh thin films (20–80 nm) grown on Al₂O₃, SiO₂/Si, and MgO, measuring $S(T, B)$, $\tau(T, B)$, and $N(T, B)$ and relating them to strain and crystal quality (XRD). In particular, we show that the bell-shaped temperature dependence of the Thomson coefficient can be adjusted by varying the film thickness, while its integral over temperature remains comparable for films grown on the same substrate. In contrast, when the substrate is changed, the crystallinity and electronic transport of the films are modified, resulting in different baseline Seebeck coefficients in the AFM and FM phases and a corresponding change in the integrated Thomson response. We show that thin films exhibit a broad temperature window of finite Thomson response, in contrast to the giant but ultranarrow response characteristic of bulk FeRh. Additionally, using experimental data and an isotropic single-band model with spin-dependent mobilities in the AFM phase, we account for the negative sign of N/B observed in FeRh/Al₂O₃ and its contrast with the larger FM-phase response.

Results and discussion

Thermoelectric properties of the FeRh/Al₂O₃ thin film

The crystal structure of the AFM and FM phases, along with their corresponding DOS per formula unit (f.u.), is shown in Fig. 1(a–d). For the FM phase, the FM DOS exhibits a pronounced imbalance, with the spin-down channel dominating at E_F . The spin-polarized DOS obtained using different functionals (see Supplementary Figure S5) shows good consistency with the PBE result, particularly near the Fermi level. In contrast, in the AFM phase, the influence of different functionals on DOS (see Supplementary Figure S6) is more pronounced, and the total DOS varies significantly from 0.35 states/eV/f.u. to 0.8 states/eV/f.u. at the Fermi level, depending on the functionals used. In this phase, spin-up and spin-down channels contribute nearly symmetrically to the DOS [Fig. 1(d)], consistent with the compensated magnetic order. Using PBE, the DOS at the Fermi level increases from ~ 0.8 states/eV/f.u. in the AFM phase to ~ 2.05 states/eV/f.u. in the FM phase. This significant enhancement in the available electronic states at the Fermi level is consistent with our experimental observation of a reduced resistivity (larger carrier density) and a more metallic Seebeck response in the FM state, as discussed below.

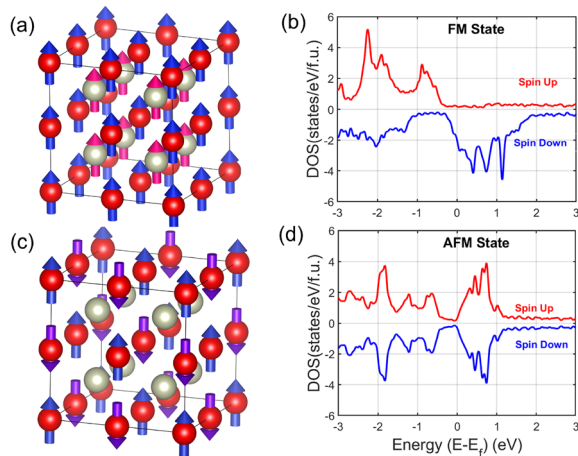


Figure 1: Chemically ordered phase with B2 (CsCl)-type crystal structure [space group 221 ($Pm\bar{3}m$)] illustrating the magnetic configuration and density of states of the FM (a-b) & AFM (c-d) phase, respectively. Iron and Rhodium atoms are shown in red and silver, respectively.

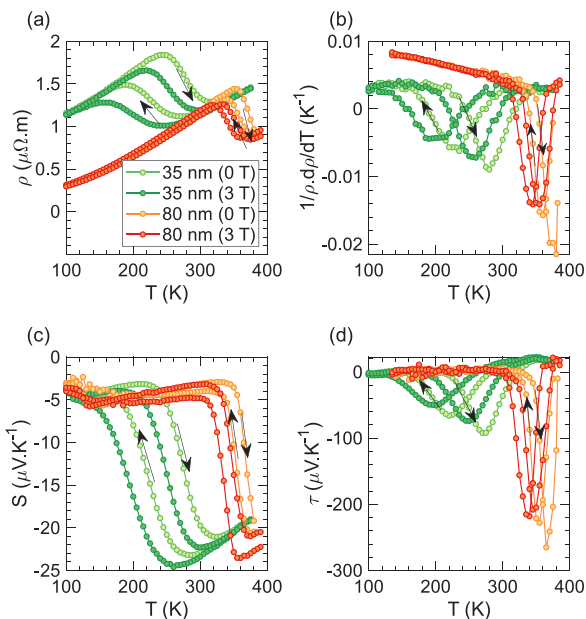


Figure 2: Temperature-dependent transport properties of FeRh/Al₂O₃ films including (a) resistivity, (b) inverse of resistivity multiplied by temperature derivative of resistivity, (c) Seebeck coefficient, and (d) Thomson coefficient. The magnetic field dependence of the thermal hysteresis is shown by different colors as indicated in the legend of (a). The black arrows indicate the temperature sweep in the upward and downward direction, respectively, around the phase transition.

The temperature dependence of resistivity (ρ) and its temperature derivative ($d\rho/dT$) is shown in Fig. 2(a-b) for 35 nm and 80 nm thick FeRh/Al₂O₃ films. A hysteretic behavior accompanied by a steep drop in ρ during heating and cooling cycles reflects the AFM–FM-phase transition, consistent with the enhanced density of states and carrier density in the FM

phase. The thinner films exhibit larger resistivity, implying an increased number of electron-scattering centers, which can be attributed to larger strain in these films as well as boundary scattering due to the size effect. It has been shown that lattice strain prevents the formation of chemically ordered phases, leading to reduced ordering in thinner FeRh films [52]. Additionally, lattice strain promotes the formation of crystalline imperfections, such as dislocations [52], which can serve as scattering centers and increase resistivity.

Upon applying a magnetic field along the [001] direction in systematic 1 T increments from 0 to 3 T, the resistivity hysteresis loop shifts toward lower temperatures [Fig. 2(a) and Supplementary Figure S8(a)]. During a slow temperature sweep across the transition, the AFM phase gradually transforms to the FM phase as the energy barrier is overcome. An external magnetic field lowers this barrier and accelerates the alignment process, thereby shifting the entire hysteresis loop to lower temperatures [53]. This field-driven shift of the AFM–FM transition is a well-established characteristic of FeRh thin films and bulk alloys, and here, it serves as a baseline for interpreting our transport and thermoelectric data. More generally, we observe that the resistivity hysteresis shifts to lower temperature under an applied field, and to higher temperature with increasing film thickness, consistently across all our samples.

Like electrical conductivity, the Seebeck coefficient is an electronic property that depends on the details of the band structure and scattering rates. During the phase transition, the magnetic and crystal structure of FeRh changes, driven by the magnetic field and temperature changes. These magnetostructural changes are accompanied by modifications in the electronic band structure and phonon dispersion. In particular, an increase in DOS and its energy derivative has been observed in the FM phase and is shown in Fig. 1(b). Consequently, the logarithmic slope of DOS at the Fermi level is larger in the FM phase [Supplementary Figure S7], which, according to the Mott relation, leads to a larger Seebeck coefficient in the FM phase [54]. To be more quantitative, we observed that S is approximately six times larger in the FM phase compared to the AFM phase [Fig. 2(c)], consistent with the theoretical calculation.

An alternative explanation for this enhancement stems from the increased entropy during the transition. In metals, S is a measure of entropy per charge carrier. Therefore, the increase in S serves as evidence that the electronic entropy increases as the system transitions to the FM phase. Independent calorimetric studies report a total entropy change of 12–19 J kg^{−1} K^{−1} across the AFM–FM transition [47, 55, 56], consistent with this interpretation. The overall entropy change across the transition has electronic, lattice (phononic), and magnetic contributions. The increase in phononic entropy is attributed to the softening of the phonons in the FM phase [57]. Cook et al. separated

the individual contributions using low-temperature specific heat, sound velocity, and photoemission measurements. They reported an electronic entropy change of $8 \pm 1 \text{ J.kg}^{-1}.\text{K}^{-1}$ [55]. Similarly, Pérez et al. found an electronic entropy change of about $8.9 \text{ J.kg}^{-1}.\text{K}^{-1}$ in Ni-doped FeRh bulk polycrystals, corresponding to a Seebeck change of $\Delta S \approx 40 \mu\text{V.K}^{-1}$ during the phase transition [56]. In comparison, our undoped thin films exhibit a smaller change of $\Delta S \approx 25 \mu\text{V.K}^{-1}$. We also note that the Seebeck coefficient of the thicker 80 nm film is slightly smaller in the FM phase compared to the 35 nm film. This is consistent with the lower resistivity observation in this thicker film.

The upper and lower temperature bounds of the Seebeck hysteresis loop are 85 K and 360 K, respectively, for the 35 nm film at 0 T, a range that is noticeably broader than that observed in the resistivity hysteresis. Similarly, the Seebeck hysteresis loop of the 80 nm film begins at a lower temperature than its resistivity hysteresis loop. The larger magnitude change and broader loop in S compared to ρ underscore the superior sensitivity of Seebeck measurements in detecting phase transitions with minimal electronic contrast, such as the AFM–FM metal–metal transition. A similar observation was made by Akhanda et al. in $1\text{T}'\text{-MoTe}_2$ single crystals [58]. In FeRh, resistivity changes remain modest because both AFM and FM phases are metallic: although the carrier concentration is larger in the FM state, the electron mean free path (MFP) is reduced, leading to only a limited net change in ρ [48, 56]. By contrast, the Seebeck coefficient is insensitive to the MFP but directly reflects changes in the density of states near the chemical potential.

Due to the metallic nature of FeRh films, the magnitude of S is much smaller than that of conventional thermoelectric materials [59–61]. Nevertheless, the abrupt changes in S across the AFM–FM transition (within the hysteresis loop) yield a large Thomson coefficient τ . A device exploiting the Thomson effect in the vicinity of the phase-change temperature could provide an alternative route for thermal-to-electrical energy conversion. The Thomson effect manifests as a reversible heat absorption or release when a current is passed through a material in the presence of a temperature gradient [62, 63].

Devices based on this effect require only a single material, in contrast to conventional Seebeck-based thermoelectric modules that need both n - and p -type legs [64]. The Thomson coefficient can be expressed as the product of the material's average temperature (T) and the temperature derivative of the Seebeck coefficient, $\tau = T.dS/dT$. The Thomson figure of merit is a combination of the thermoelectric figure of merit and the Thomson to Seebeck coefficient ratio [65]. τ is generally small in most materials; consequently, the phase transition-induced sharp change in the S has fueled the exploration of Thomson effect-based devices using phase-change materials. This was highlighted in bulk FeRh recently, where a

very large τ was reported over an extremely narrow temperature range with almost zero values outside of this range [48].

For practical cooling or power generation, τ must remain large (on the order of $10^2 \mu\text{V.K}^{-1}$) across a wide temperature span. The temperature-dependent τ of FeRh/ Al_2O_3 , extracted from the measured $S(T)$, is shown in Fig. 2(d). Both heating and cooling sweeps reveal distinct peaks, reflecting the hysteresis in S . The Thomson coefficient peak observed in all FeRh films studied here within the hysteresis loop is larger than the Seebeck coefficient. Importantly, while the peak τ values in our thin films are smaller than those in bulk FeRh, they persist over a much broader range, making them more suitable for device applications. For the 35 nm film, S reaches peaks of $-21 \mu\text{V.K}^{-1}$ (heating) and $-23 \mu\text{V.K}^{-1}$ (cooling), whereas τ attains $-98 \mu\text{V.K}^{-1}$ and $-66 \mu\text{V.K}^{-1}$, respectively. In the 80 nm film, the peak τ exceeds $-250 \mu\text{V.K}^{-1}$. In contrast to bulk FeRh, which exhibits extremely large but sharply localized Thomson peaks [48], our thin-film samples reveal a trade-off between peak magnitude and operational window, with smaller maxima but a far broader temperature range that is more relevant for practical device applications. The nonzero τ spans 135–370 K for the 35 nm film and 305–400 K for the 80 nm film, extending the operational window by more than an order of magnitude relative to bulk reports. This behavior reflects the peak–window trade-off described above.

Since we only measured transport properties and not thermodynamic properties, it is not possible to accurately estimate the phase transition temperature, T_c . Instead, we define an apparent transport transition temperature T_c as the temperature where $|\tau|$ reaches its maximum. For clarity, we also introduce the notation T_{low} and T_{high} to denote the temperatures at which the heating and cooling branches of the $\tau(T)$ hysteresis loop intersect, representing the temperature window over which the Thomson coefficient is non-negligible.

With this notation, T_{low} decreases linearly by approximately 5 K for every 1 T magnetic field increment in the 35 nm film. In contrast, T_{high} follows a non-linear pattern with approximately 0 K, 10 K, and 0 K decreases during the increments 0–1 T, 1–2 T, and 2–3 T, respectively [see Table 1]. As a result, the hysteresis span ($T_{high} - T_{low}$) varies slightly with field. For the 80 nm FeRh/ Al_2O_3 film, the apparent upper bound of ~ 380 K remains unchanged for 0, 1, and 2 T, though this value is limited by our measurement range and does not represent the true T_{high} . The lower bound decreases systematically from 305 K at 0 T to 285 K at 2 T, consistent with the field-induced stabilization of the FM phase.

Substrate dependence of the transport parameters

Next, we examine the influence of the substrate on the transport properties. The temperature-dependent variation of the

TABLE 1: Upper and lower bounds of hysteresis in the Thomson curve of the FeRh/Al₂O₃ films.

Thickness (nm)	Magnetic field (T)	T _C (cooling) (K)	T _C (heating) (K)	T _{low} (K)	T _{high} (K)
80	0	355	365	305	>380
80	1	350	365	300	>380
80	2	345	355	285	>380
80	3	340	345	280	380
35	0	220	270	135	370
35	1	220	270	130	370
35	2	210	260	125	360
35	3	200	250	120	360

The measurement is limited by the experimental setup to 100 to 380 K.

transport properties, namely the resistivity, the Seebeck coefficient, and the Thomson coefficient, of FeRh/SiO₂/Si and FeRh/MgO is shown in Fig. 3(a-i). Because the full τ hysteresis could not be captured for all samples, we use its onset temperature (T_{low}) as a proxy for the phase transition temperature. For 80 nm films under an applied field of 3 T, T_{low} is lowest for FeRh/SiO₂/Si (~100 K), followed by FeRh/Al₂O₃ (~280 K), and highest for FeRh/MgO [batch 1] (~345 K). Previous studies have reported higher T_c values in MgO-based films relative to Al₂O₃, attributed to the rougher surface morphology of Al₂O₃-grown films, [66], and our observations are consistent with this trend. Similarly, we observe that the phase transition of FeRh/MgO [batch 1] is significantly shifted to higher temperatures compared with the other films and lies beyond our measurement range. Batch 2 FeRh/MgO film in Fig. 3(g)

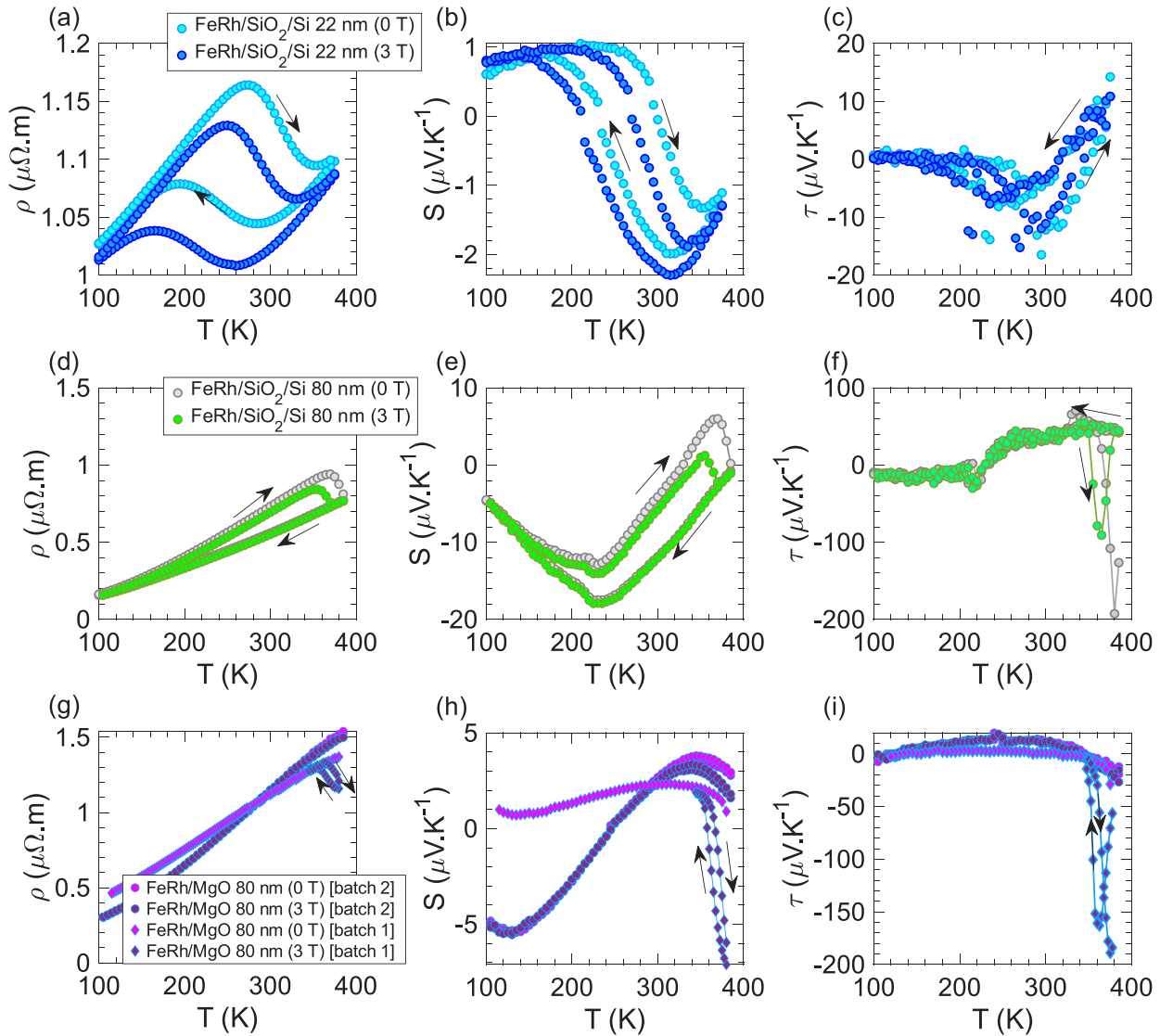


Figure 3: Variations in resistivity, Seebeck coefficient, and Thomson coefficient of FeRh/SiO₂/Si 22 nm (a-c), FeRh/SiO₂/Si 80 nm (d-f), and FeRh/MgO 80 nm (g-i) with temperature. In the case of FeRh/MgO, two films are reported and labeled as batch 1 and batch 2, as detailed in Tables 3, S1, and S2. The black arrows denote the temperature sweeping up and down around the phase transition.

does not show any hysteresis, but a small deviation at 3 T at high temperature, pointing to the start of the phase transition.

Unlike the thickness-controlled case on Al_2O_3 , changing the substrate modifies the quality and crystallinity of the films, thereby altering the electronic transport itself and preventing a simple reshaping of the Thomson response at a fixed baseline Seebeck. Different substrates induce varying degrees of strain on the films, which can significantly affect T_c . For example, it is shown that when FeRh films are deposited on an MgO substrate, the lattice mismatch generates strain, altering the crystal structure's c/a ratio [46]. Studies show that even a 1% change in this ratio can significantly impact the transition temperature, shifting it by as much as 40 K [67]. Upon cooling the samples following FeRh deposition, a coefficient of thermal expansion (CTE) mismatch between the FeRh film and the substrate places the film in an in-plane stress state. In addition to structural distortion, the magnetic properties of FeRh can also be affected by strain-induced changes in electronic structure. Strain can lead to a reorientation of the Fe moments, altering the AFM to FM transition temperature, as evidenced by Mössbauer spectroscopy [68].

A clear observation of the effect of strain is the thickness dependence of T_c . Increasing the film thickness allows partial relaxation of strain, raising T_c toward the bulk value. Consistent with this expectation, we observe higher T_c values in thicker FeRh films grown on Al_2O_3 and SiO_2/Si substrates, as noted also in the previous section. In addition to thickness, the type of strain also has a significant impact on the T_c . FeRh/ Al_2O_3 films experience out-of-plane compressive and in-plane tensile strain [69], while FeRh/MgO exhibits the opposite, with out-of-plane tensile and in-plane compressive strain. Despite similar unit-cell volumes, these differences yield distinct tetragonality ratios ($c/a > 1$ for MgO, $c/a < 1$ for Al_2O_3). Quantitatively, 110 nm FeRh/ Al_2O_3 films show an in-plane tensile strain of -0.27% , while 110 nm FeRh/MgO(100) films exhibit an in-plane compressive strain of $+0.33\%$ [70]. Such differences produce large shifts in T_c [71], with changes of more than 60 K reported when in-plane stress is switched from compressive to tensile [72].

We experimentally examined the strain in the 80 nm films, and the results are summarized in Table 2. Upon cooling, a greater substrate CTE results in biaxial compressive strain in the film. This in-plane strain confines the lattice, corresponding to an out-of-plane expansion, observed as a positive strain in the FeRh deposited on MgO. In contrast, a smaller substrate CTE results in a film tensile strain in-plane, a relative compression of the lattice out-of-plane, and a negative strain, as seen in the FeRh films deposited on Al_2O_3 and SiO_2/Si with the latter representing a much larger strain.

X-ray diffraction patterns of FeRh films deposited onto (0001) Al_2O_3 , 1 μm amorphous SiO_2 on (001) Si, and (001)

MgO are shown in Fig. 4(a-c), respectively. Both FeRh and substrate reflections are indexed by dashed lines. Multiple FeRh reflections are present in Fig. 4(a) FeRh/ Al_2O_3 , indicative of film polycrystallinity, with the intense (111) FeRh reflection characteristic of preferred (111) out-of-plane texture. Similar FeRh polycrystallinity of films on SiO_2/Si is present in Fig. 4(b), with a preferred (100) FeRh texture. The superposition of MgO and FeRh reflections, seen in Fig. 4(c), is characteristic of epitaxial growth of FeRh onto the MgO substrate.

In addition to the shift in T_c , we also observe changes in the sharpness of the transition. Specifically, all films studied here exhibit a broader transition compared to bulk FeRh [48]. This broadening has been attributed to defect-driven phenomena, as discussed by Keavney et al.. Their study of the evolution of average lattice expansion, using spatially resolved diffraction during the phase transition, revealed a heterogeneous mixture of AFM and FM phases in the films. These phases appear as nucleated islands that form and merge into larger domains at higher or lower temperatures, depending on the sweep direction. Regions of the film near the defects experience a lower effective transition temperature as a result of localized strain and/or local changes in the electronic structure. The distribution of nucleation temperatures arises from defects of different types, which contribute to a slower overall transition. Thus, films with fewer defects should exhibit a steeper phase transition.

Among the films, the FeRh/ SiO_2/Si exhibits the broadest phase transition, as evidenced in Fig. 3. Consistent with this behavior, the films on SiO_2/Si are polycrystalline and display the largest tensile strain [Fig. 4(b), Table 2]. In contrast, FeRh grown on MgO has a minimal lattice mismatch of only $\sim 0.3\%$ relative to the FCC cubic lattice of MgO ($a = 4.212 \text{ \AA}$), with a 45° in-plane rotation that demonstrates epitaxial matching [Fig. 4(c)]. This epitaxial growth produces in-plane compressive strain (Table 2), which shifts the transition temperature to higher values.

In addition to the shift in T_c , strain appears to have a complex and unpredictable influence on the sign of the Seebeck coefficient, particularly in the AFM phase. In the FM phase, the Fermi level is deep in the band, and the slope of DOS is positive at the Fermi level [Fig. 1, Figure S7], resulting in an electron-like behavior and a negative Seebeck coefficient for all three films. Consequently, bulk [48] as well as thin films (this work) always demonstrate a negative Seebeck coefficient in this phase. By contrast, in the AFM state, the Fermi level is very close to a minimum in the DOS as shown in Fig. 1, where strain or doping can shift it toward positive or negative slopes [Fig. S11]. This makes the AFM-phase Seebeck coefficient highly sensitive to multiple competing parameters. Bulk FeRh has shown an almost zero Seebeck coefficient [48], whereas Ni-doped FeRh demonstrates a positive Seebeck coefficient. In our films, substrate and microstructure lead to different behaviors: FeRh/ Al_2

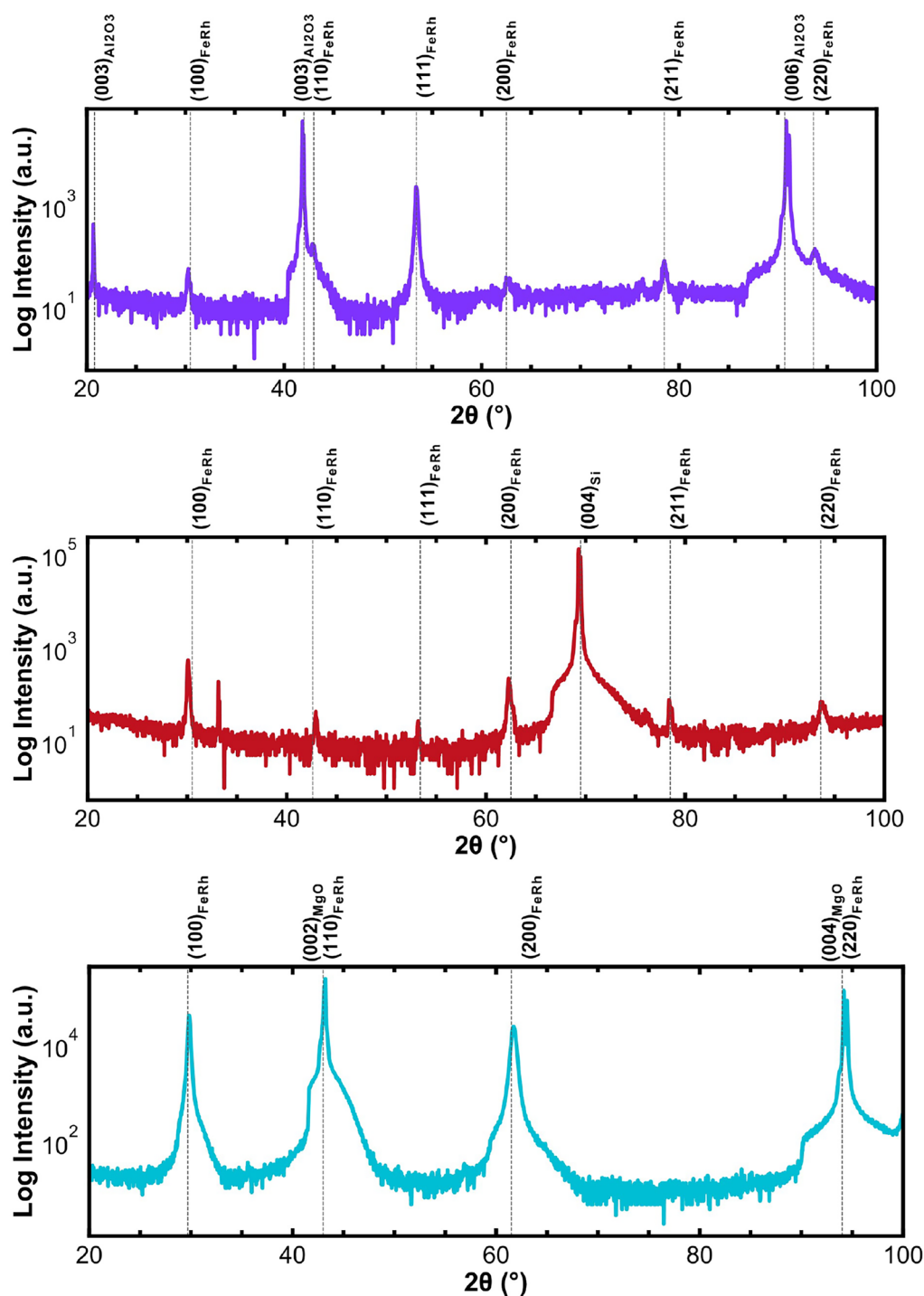


Figure 4: Bragg–Brentano X-ray diffraction patterns of batch 2 FeRh films deposited on (a) (0001) Al_2O_3 , (b) SiO_2 /(001)-oriented Si, and (c) (001) MgO substrates.

TABLE 2: Out-of-plane strain analysis of the 80 nm films (batch 2) under zero magnetic field.

Substrate	FeRh Reflection	Strain	T_C (cooling)	T_C (heating)	T_{low}	T_{high}
(0001) Al_2O_3	(111)	-1.079×10^{-4}	355 K	365 K	305 K	> 380 K
SiO_2 /(001)Si	(220)	-1.367×10^{-2}	215 K	380 K	120 K	> 380 K
(001) MgO	(200)	6.772×10^{-3}	UR	UR	UR	UR

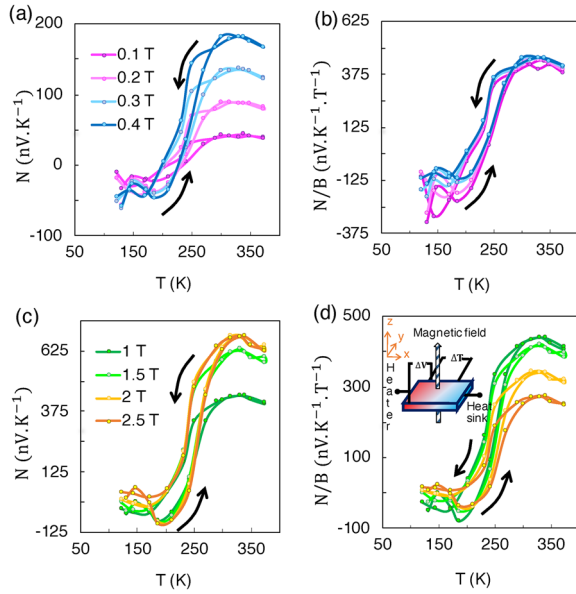


Figure 5: The temperature and magnetic field dependence of the Nernst thermopower (N) and the Nernst coefficient (N/B) of 35 nm FeRh/Al₂O₃. The legends in (b) and (d) align with those in (a) and (c), respectively. The black arrows indicate temperature sweeping up and down around the phase transition. The inset of (d) depicts the Nernst coefficient measurement setup. For improved readability, graphs corresponding to magnetic fields below and above 1 T are presented in separate panels.

O₃ films display negative S in the AFM phase, while FeRh/SiO₂/Si films show more varied responses—the thinner film exhibits a positive S , whereas the thicker film shows a negative S that reverses sign within the mixed-phase region before returning to negative in the FM state. The batch 2 FeRh film on MgO has a negative S in the AFM phase, but when it is aged (oxidized on the surface), it has a positive Seebeck coefficient. Since the Seebeck value is small in general, these variations are expected. These findings highlight the substrate- and strain-dependent tunability of transport in FeRh films.

Nernst effect in FeRh films

While the Thomson effect is the primary focus of this work, Nernst measurements provide an additional probe of magneto-thermoelectric transport across the AFM–FM transition. The temperature dependence of the Nernst thermopower (N) and Nernst coefficient (N/B) is plotted in Fig. 5(a–d) for the 35 nm films. For the 80 nm film, as noted earlier, the higher phase transition temperature limits the accessible hysteresis window due to instrument constraints. In both cases, we observe a narrower but similarly shaped hysteresis compared to S . The absolute value of N/B is larger in the FM phase than in the AFM phase. The FM phase's resistivity is lower than that of the AFM phase due to the larger carrier concentration of the FM phase. However, the carrier mobility is smaller in the FM phase due to stronger electron–phonon and

electron–electron-scattering rates. Therefore, the larger Nernst coefficient observed in the FM phase is counter-intuitive since the Nernst coefficient tends to be larger at lower temperatures and higher carrier mobility [59]. Furthermore, at a given temperature, the FM-phase N/B remains constant and field-independent up to ~ 0.4 T, while at higher fields, it decreases, indicating that higher-order contributions become relevant and that the Nernst voltage is no longer linear in B . The Nernst coefficient is about an order of magnitude smaller than the Seebeck coefficient in these samples. Importantly, the anomalous contribution at zero field is negligible in both FM and AFM states [51, 73], consistent with the small out-of-plane magnetization of these films.

In the FM phase, where spin-down carriers dominate [Fig. 1(b)], the behavior of the ordinary Nernst effect can be described using a single-band model [74]. For isotropic bands, the isothermal Nernst thermopower is given by

$$N = S \left(\frac{r}{r + 1.5} \right) \mu_H B_z, \quad (1)$$

where S is the Seebeck coefficient, μ_H is the Hall mobility, and r characterizes the energy dependence of the relaxation time $\tau_f \propto E^r$. B_z is the internal magnetic field, equal to the applied field plus the contribution from the internal magnetization. Magnetometry shows that up to 3 T and at 350 K, this magnetization increases the effective internal field by a factor of ~ 2.2 (Supplementary Figure S12 and Ref. [73]). Thus, the effective Nernst coefficient relative to the external field is

$$\frac{N}{B_{z,\text{ext}}} = 2.2S \left(\frac{r}{r + 1.5} \right) \mu_H. \quad (2)$$

Combining results from Perez et al. [56] and Modak et al. [48], and considering $\mu_H = 40 \text{ cm}^2/\text{V.s}$, $S = -25 \text{ } \mu\text{V.K}^{-1}$ (measured value of Seebeck coefficient at 300 K from this work, see Fig. 2), and $r = -0.8$, we obtain $N/B = 228 \text{ nV.K}^{-1}.\text{T}^{-1}$, which is close to the experimentally measured value of approximately $375 \text{ nV.K}^{-1}.\text{T}^{-1}$ in Fig. 5 considering the simplicity of the model. Here, we did not use any adjusting parameters. The r value is obtained by fitting the FM DOS close to the Fermi level with the power law and assuming $\tau_f \propto 1/\text{DOS}$.

The AFM phase is more complex, with two parallel currents from spin-up and spin-down contributing almost equally. Because the Fermi level lies near a DOS minimum [Supplementary Figure S6], strain-induced shifts of E_F complicate fitting with a simple power law. In the mixed-phase region, the presence of the ferromagnetic regions results in spin-dependent scattering rates and unequal mobilities for the two types of spins, which then results in terms in the Nernst coefficient that have contributions from both bands (mixed terms) [74]. The sign of the Nernst thermopower in this region then depends on the details of the mobility of spin-up and spin-down carriers and can be obtained using:

$$N = \frac{\sigma_0^{\uparrow 2} N_0^{\uparrow} + \sigma_0^{\downarrow 2} N_0^{\downarrow}}{(\sigma_0^{\uparrow} + \sigma_0^{\downarrow})^2} + \frac{1}{T^2} \frac{\sigma_0^{\uparrow} \beta_0^{\downarrow} \mu_{\beta}^{\downarrow} - \sigma_0^{\downarrow} \beta_0^{\uparrow} \mu_H^{\downarrow} + \sigma_0^{\downarrow} \beta_0^{\uparrow} \mu_{\beta}^{\uparrow} - \sigma_0^{\uparrow} \beta_0^{\downarrow} \mu_H^{\uparrow}}{(\sigma_0^{\uparrow} + \sigma_0^{\downarrow})^2} B_z \quad (3)$$

where $N_0^{\uparrow, \downarrow}$ and $\sigma_0^{\uparrow, \downarrow}$ denote the Nernst coefficients and conductivities of spin-up and spin-down channels, respectively, and μ_{β} and β_0 are thermal-mobility and thermoelectric response functions as defined in Ref. [74]. The first term represents a weighted average of the spin-resolved Nernst coefficients. The second term is the mixed term and contains contributions from both spin-up and spin-down channels. The sign of N in this regime is therefore governed by the relative mobilities of the spin-up and spin-down channels.

While previous studies of FeRh films have emphasized the anomalous Nernst effect [51, 73], our results provide a quantitative account of the ordinary Nernst effect in the FM phase, where a simple single-band model reproduces both the sign and the magnitude without adjustable parameters. In the AFM phase, the situation is more complex: our two-current framework identifies the dependence of the Nernst sign on the relative mobilities of spin-up and spin-down channels, even though a full quantitative description is not possible without fitting parameters.

Conclusion

We investigated the thickness- and substrate-dependent Thomson response of near-equimolar FeRh thin films across the first-order AFM–FM transition near room temperature. By measuring $S(T, B)$, $\tau(T, B)$, and $N(T, B)$ for films with thicknesses of 20–80 nm grown on Al_2O_3 , SiO_2/Si , and MgO, we establish that the Thomson coefficient provides a particularly sensitive fingerprint of how the transition is manifested in electronic transport. Full characterization of the hysteresis loop for MgO films requires extension to higher temperatures, which is currently beyond the capabilities of our equipment.

For films grown on the same substrate (Al_2O_3), varying the film thickness systematically reshapes the temperature dependence of $\tau(T)$, revealing a clear trade-off between the peak magnitude of the Thomson coefficient and the temperature window over which it remains finite: thicker films exhibit larger peak values over narrower ranges, whereas thinner films maintain smaller peaks across substantially broader temperature ranges. In this thickness-controlled regime, the temperature-integrated Thomson response remains comparable, consistent with a redistribution of the transition-associated transport signature over temperature. In contrast, changing the substrate modifies film crystallinity and electronic transport in the AFM and FM

phases, leading to substantial variations in the baseline Seebeck coefficient and a corresponding qualitative change in $\tau(T)$ and its integrated response. This shows that thin films can exhibit a broad temperature window of finite Thomson response, in sharp contrast to the giant but ultranarrow behavior reported for bulk FeRh, highlighting thin-film growth as a practical route to engineering both the magnitude and operational temperature span of the Thomson effect in magnetic materials.

Finally, we report the Nernst effect in FeRh/ Al_2O_3 films. The measured N is two orders of magnitude smaller than S , displays a clear hysteresis with a shape similar to that of S , and changes sign during the phase transition. Quantitative agreement with a single-band model is achieved in the FM phase, while in the AFM phase, the sign of N is governed by the relative mobilities of spin-up and spin-down channels. Despite higher mobility in the AFM state, the FM phase shows larger $|N|$ due to the contribution from its internal magnetization. To further elucidate these observations, future work could include in-plane characterization of the strain, as well as microscopic structural characterization at varying temperatures. This would help clarify the roles of microstructure and disorder in shaping the thermomagnetic response.

Materials and methods

Sample preparation

For all FeRh films used in this study, magnetron sputtering was chosen as the growth method. Three substrates were used: (0001) Al_2O_3 , (001) MgO, and 1 μm amorphous SiO_2 deposited on (001) Si via low-pressure chemical vapor deposition (LPCVD). MgO has previously been used [75] as a substrate for FeRh growth due to a suitable lattice match for high-quality epitaxial growth. FeRh films on Al_2O_3 have been shown to be nonepitaxial, with rougher surface morphology compared to MgO films [66]. The substrate plays a crucial role in the manifestation of a magnetostructural transition within FeRh [76–78].

During sample growth, an AJA International Orion II magnetron sputter system was employed with a 4-inch Inconel backlight bulb heated substrate holder raised to 630°C substrate temperature. Before sample growth, the substrates were cleaned of organic contaminants in 10 mTorr Ar by a 10 W RF back-sputter. Growths were conducted from a stoichiometric FeRh sintered 2-inch target, mounted on a 2-inch magnetron configured for magnetic targets, gradually raised to a DC power of 75 W in an Ar working gas pressure of 4 mTorr. Films with thicknesses in the 20–80 nm range were grown and verified by X-ray reflectivity (XRR) and then analyzed using a fast Fourier transform. After growth, the films were annealed at an elevated temperature of 730°C for 1 h under a high vacuum ($\sim 4 \times 10^{-8}$ Torr) before cooling to room temperature. These annealing conditions were chosen to promote B2 chemical ordering and

TABLE 3: Properties of FeRh films studied in this work.

Batch	Thickness	Substrate	Fe Atom%
Batch 1	35 nm	Al ₂ O ₃	53.04
	80 nm	MgO	52.50
	22 nm	SiO ₂ /Si	52.00
	80 nm	Al ₂ O ₃	50.79
Batch 2	80 nm	MgO	50.90
	80 nm	SiO ₂ /Si	50.96

improve crystallinity. Details on structural analysis can be found in the supplementary material (Figs. S1, S2, and S3). Table 3 summarizes the properties of the films.

Strain analysis

A Bruker D2 phaser with a Cu-K α source was used to collect X-ray diffraction patterns in a Bragg–Brentano geometry from the 20–100° 2θ range. Peaks within the patterns were fitted using Pearson-VII peak shapes in the LIPRAS software [79]. After extracting substrate d -spacings from LIPRAS fit data [79], sample displacement error was corrected using the following equation [80]:

$$\frac{\Delta d}{d} = -\frac{D \cos^2(\theta)}{R \sin(\theta)} \quad (4)$$

where d is reported d -spacing, Δd is the change in d -spacing, D is the specimen displacement parallel to the diffraction vector, R is the diffractometer radius, and θ is the angle of diffraction. Reported d -spacings for Al₂O₃ [81], Si [82], MgO [83], and FeRh [84] were exported from the inorganic crystal structure database. From there, the out-of-plane strain was calculated using the difference between the reference and measured d -spacing of high-angle FeRh reflections. The analysis was performed primarily on the 80 nm films, as the signal from the thinner films was noisy and reliable analysis was not possible. It should also be noted that direct determination of in-plane strain, which would require reciprocal-space mapping or grazing-incidence measurements, was beyond the scope of this work and therefore not pursued.

Transport property measurements

The temperature-dependent Seebeck coefficient measurement was conducted within the thermal transport option of a Versalab setup from Quantum Design Inc., utilizing the one-heater two-thermometer configuration. 0.5 mm wide gold-coated flat copper wires were used to make the electrical contacts. One end of the sample was connected to a resistive heater, while the other end was clamped to the cold foot of the puck, serving as the heat sink. Between the heater and heat sink, two thermal

transport shoes, capable of measuring both temperature and voltage, were placed to measure the temperature gradient and the corresponding voltage developed. The temperature difference (ΔT) for all samples was consistently maintained below 1% of the average sample temperature. A four-point probe configuration was employed to measure the resistivity of the samples, passing current from the heater side toward the cold foot or the heat sink. In the Nernst measurement, an external magnetic field was applied perpendicular to the thermal gradient to generate a transverse voltage. The developed voltage was measured using Keithley nanovoltmeter 2182A. To facilitate the measurement, the voltage lead of the cold side shoe was separated and connected transversely opposite to the hot side shoe along the width of the samples. Estimated errors in the S , resistivity, and Nernst measurements are approximately 5%, 3%, and 10%, respectively. Major sources of error include fitting errors in the MultiVu software (for Seebeck measurement), sample size measurement errors, and inherent instrumental errors.

Computational methods

The spin-polarized first principle calculations of the density of states were performed using DFT as implemented in VASP code [85–87] with Projected Augmented Wave (PAW) potentials [88]. We employed different functionals to describe the effects of electron exchange and correlation effects and studied the functional-dependent stability of the system in terms of the local magnetic moment and external pressure of the system. Finally, Generalized Gradient Approximation (GGA) by Perdew Burke and Ernzerhof (PBE) [89] was adopted for further calculations. The rationale for choosing PBE functionals over other functionals, e.g., LDA [90], PBEsol [91], SCAN [92], TPSS [93], and DFT+U+J [95, 95], is discussed in Supplementary Information. The spin-orbit coupling (SOC) was also considered to complete the study but the density of states (DOS), as shown in Supplementary Information Figure S4, revealed that the effect of SOC is negligible. Similar results were also reported by other studies [96, 97]. Therefore, SOC effects were not considered for further calculations. For electronic structure calculations, we used a $2 \times 2 \times 2$ supercell comprising 16 atoms to simulate the AFM and FM phases of FeRh. According to PAW pseudopotentials, Fe(3d, 4 s) and Rh (4p, 5 s, and 4d) orbitals were treated as valence states in this analysis. The plane-wave energy cutoff of 720 eV, which is more than 2.5 times the standard value, was chosen with a self-consistency threshold of 10^{-9} eV. A dense Monkhorst-Pack k -mesh grid of $17 \times 17 \times 17$ was used for sampling the Brillouin zone. As FeRh is metallic in nature, a smearing value of 0.05 eV according to the Methfessel–Paxton scheme was used for self-consistent as well as DOS calculations. The lattice parameters for AFM and FM phases were fixed to experimental

values $a = 2.99 \text{ \AA}$ [33] and $a = 3.01 \text{ \AA}$ [13], respectively. With these parameters, the magnetic moments obtained from DFT calculations are $\mu_{Fe}=3.21$ and $\mu_{Rh}=1.05$ in the FM phase, and $\mu_{Fe}=3.12$ and $\mu_{Rh}=0$ in the AFM phase. These values are in good agreement with other calculations [97–100] and experimental [101, 102] results. The total energy per atom for the AFM phase is 0.02 eV lower than the FM phase at $T=0 \text{ K}$.

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Author contributions

Md Sabbir Akhanda: Transport Measurements, Formal Analysis, and Writing-original draft. Sree Sourav Das: First-Principles Calculations, Writing - Review & Editing. Megan Lenox and Benjamin Aronson: XRD and Strain Analysis. Shelby Fields and Steven P Bennett: Thin-film growth. Jae Won Cho and Jung-Min Cho: Magnetization measurements. Bellave Shivaram: Conceptualization, Writing - Review & Editing. Sang-Kwon Lee: Resources, Formal analysis, Writing - Review & Editing, Funding acquisition. Jon Ihlefeld: Strain analysis, Writing - Review & Editing. Mona Zebarjadi: Conceptualization, Validation, Formal analysis, Resources, Writing - Review & Editing, Supervision, Funding acquisition.

Data availability

The data supporting this article have been included as part of the Supplementary Information.

Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Supplementary Information

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