

Emerging magnetic phase in the orthoferrite HoFeO₃ detected by spin Hall magnetoresistance and spin Seebeck effect

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The orthoferrite HoFeO₃ exhibits a complex interplay of magnetic interactions between the antiferromagnetically ordered Fe³⁺ spins and the rare-earth Ho³⁺ ions. In this work, we combine superconducting quantum interference device (SQUID) magnetometry studies with electrical spin transport measurements of the spin Hall magnetoresistance (SMR) and spin Seebeck effect (SSE) on a HoFeO₃ single crystal interfaced by a thin Pt layer. Our measurements show that the increase in Ho magnetic susceptibility below the Morin-like spin reorientation transition at 58 K affects the magnetic anisotropy of the Fe sublattice and induces a tilting of the easy axis. Below the ordering temperature of Ho at 4 K, we observe a rapid increase in the SSE signal associated with the emergence of low-energy magnon modes with the onset of Ho-Ho exchange. These findings demonstrate that spin transport techniques can sensitively probe rare-earth-induced modifications of anisotropy and spin dynamics in complex oxides, opening pathways to electrically access low-energy 4f excitations.

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I. INTRODUCTION

Rare-earth orthoferrites have emerged as a promising platform for spintronic devices due to their unique combination of antiferromagnetic and magnetoelectric coupling, as well as tunable spin dynamics. These materials can host ultrafast magnetic switching [1–3], multiferroic behavior [4–8], and complex spin textures, making them valuable candidates for both fundamental studies and device engineering.

In the family of orthoferrites, HoFeO₃ hosts two magnetically active sublattices, Fe³⁺ and Ho³⁺. The interplay between antiferromagnetic ordered Fe³⁺ spins and highly anisotropic Ho³⁺ moments gives rise to a rich magnetic phase diagram tunable via field and strain [9]. The exchange coupling between the two sublattices, combined with structural distortions, determines the magnetic anisotropy [10–12], making HoFeO₃ a model system for investigating exchange interactions and spin anisotropy in rare-earth transition metal systems [13,14].

HoFeO₃ has a distorted perovskite structure of the Pbnm space group. The unit cell contains four Ho³⁺ ions, four Fe³⁺ ions, and twelve O²⁻ ions [Fig. 1(a)]. The Fe³⁺ spins ($S = 5/2$) order antiferromagnetically below the Néel temperature $T_N \approx 645$ K [15]. Due to the lack of inversion symmetry, a nonzero Dzyaloshinskii-Moriya interaction

(DMI) term arises around certain Fe-O-Fe bonds, introducing a small canting angle ($\approx 0.5^\circ$) between the spins of neighboring Fe ions and giving rise to a weak uncompensated moment $\mathbf{m}_{\text{Fe}}$ [12]. The Ho³⁺ ions have a high total angular momentum $\mathbf{J} = 8$. At temperatures below ~ 4 K, the Ho³⁺ ions become magnetically ordered with the onset of a negative Ho-Ho exchange [16].

The temperature-dependent anisotropy in Fe-Ho exchange associated with structural distortions gives rise to several spin reorientation transitions in the temperature range 38 K–58 K [Fig. 1(c)]. Below 38 K, HoFeO₃ resides in the Γ_2 magnetic phase, where the Néel vector ℓ_{Fe} lies along the c axis of the crystal, and the weak ferromagnetic moment $\mathbf{m}_{\text{Fe}}$ lies along the a axis. As the temperature increases above 38 K, ℓ_{Fe} begins a continuous rotation within the bc plane, gradually tilting away from the c axis while $\mathbf{m}_{\text{Fe}}$ remains pinned along the a axis—a regime known as the Γ_{12} phase. At 52 K, the system undergoes a first-order transition into the Γ_{24} phase: here, ℓ_{Fe} abruptly reorients into the ac plane making an angle of approximately 15° – 20° with the c axis, while $\mathbf{m}_{\text{Fe}}$ tilts correspondingly away from the a -axis into the ac -plane. Between 52 K and 58 K, ℓ_{Fe} continues its smooth rotation toward the a axis, culminating at 58 K in the Γ_4 phase, where ℓ_{Fe} stabilizes along the a axis and $\mathbf{m}_{\text{Fe}}$ along the c axis [12,17,18].

In this work, we use a combination of superconducting quantum interference device (SQUID) magnetometry and electrical spin transport measurements, including spin Hall magnetoresistance (SMR) and spin Seebeck effect (SSE), to study how 3d-4f exchange between the Fe and Ho sublattices affects the low-temperature anisotropy of HoFeO₃ below 38 K and find a rare-earth-induced tilting of the anisotropy axis

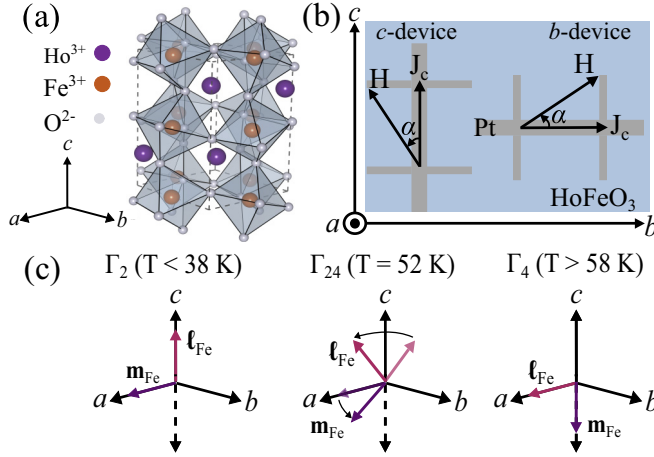


FIG. 1. (a) Crystallographic unit cell of HoFeO_3 (Pbnm), illustrating the positions of Ho^{3+} (purple), Fe^{3+} (orange), and O^{2-} (gray) ions. The Fe^{3+} ions occupy the centers of corner-sharing oxygen octahedra, while Ho^{3+} ions sit between the FeO_6 frameworks. (b) Schematic of the two Hall bar devices used for SMR and SSE measurements. An alternating current $\mathbf{J}_c$ is applied along the bar, and longitudinal and transverse voltages are measured. The external magnetic field is rotated in the bc plane, where the angle α is defined relative to the current direction. (c) Schematic representation of the spin reorienting transitions of the Fe^{3+} sublattice in HoFeO_3 .

and the emergence of low-energy magnon excitations with the onset of spontaneous magnetic ordering in the Ho sublattice.

II. EXPERIMENTAL METHODS

a -cut HoFeO_3 single crystals were grown by the floating zone method. A 4 nm thick Pt layer was sputtered on the polished bc surface, after light milling. Prior to Pt deposition, the surface of the single crystal was cleaned *in situ* using a soft Ar etch at 50 W on a 4-inch substrate holder at 10 m Torr. A 4 nm thick Pt layer was then sputtered at 3 m Torr with a typical deposition rate of $0.5 \text{ \AA/s}$ at room temperature. Hall bars were patterned in the Pt layer via optical lithography and argon etching [Fig. 1(b)], with the long axis parallel to the b axis (b device) or the c axis (c device). The width of the bars is $50 \text{ \mu m}$, while the longitudinal contacts is $300 \text{ \mu m}$.

SMR and SSE measurements were performed via first- and second-harmonic lock-in detection, respectively, with an AC applied at 77 Hz along the long axis of the bar. For the first-harmonic SMR measurements, a current of $I_{AC} = 200 \text{ \mu A}$ was used. In contrast, for the second-harmonic SSE measurements, a higher current of $I_{AC} = 1 \text{ mA}$ was applied to increase the temperature gradient at the interface between Pt and HoFeO_3 .

III. RESULTS

We start by presenting magnetometry studies of the HoFeO_3 single crystal.

Figure 2 shows SQUID magnetization measurements as a function of temperature for field applied along different directions with respect to the HoFeO_3 crystal axes. The spin-reorientation transition from the Γ_4 to the Γ_2 phase is clearly visible between 58 K and 52 K in the low field (0.01 T)

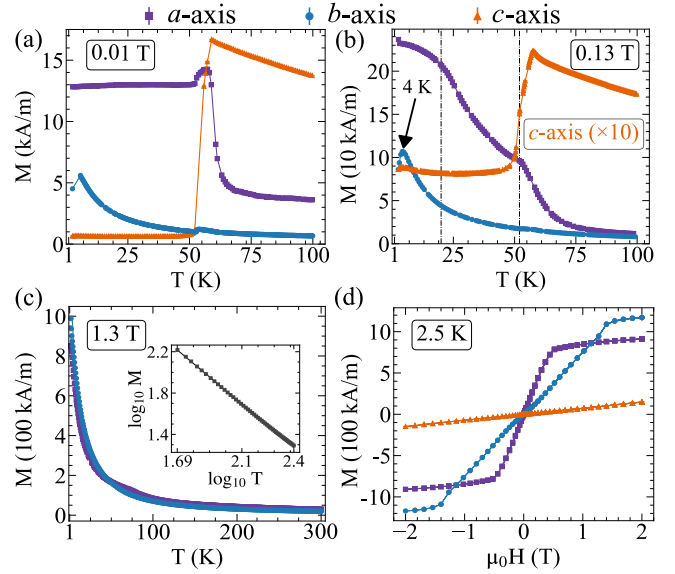


FIG. 2. Temperature-dependent magnetization of HoFeO_3 measured via SQUID magnetometry for a magnetic field of 0.01 T (a) and 0.13 T (b), applied along the out-of-plane a axis and in-plane b and c axes. In (b), the c -axis trace is rescaled by a factor of ten, as also indicated on the plot. (c) Magnetization curves measured at 1.3 T along the a and b axes, fitted using a Brillouin model. The inset shows the log-log plot fitted with a Curie-like power law. (d) Field-dependent magnetization measured at 2.5 K for fields applied along all three crystallographic directions, revealing anisotropic saturation behavior. All measurements were carried out under zero-field-cooled (ZFC) conditions.

magnetization curves along the c and a axes [Fig. 2(a)]. During this transition, $\mathbf{m}_{\text{Fe}}$ rotates from the c axis to the a axis, while $\mathbf{l}_{\text{Fe}}$ rotates from the a axis to the c axis.

The distinct kink near 4 K observed in the b and c sweep [Fig. 2(b)] corresponds to the onset of long-range antiferromagnetic ordering in the Ho sublattice. This anomaly has also been observed in heat capacity [19,20] and Mössbauer spectroscopy studies [21], where it was likewise attributed to spontaneous magnetic ordering of the Ho^{3+} moments. Neutron diffraction and Mössbauer studies further establish that the ordered Ho^{3+} moments lie in the ab plane, tilted by approximately 27° from the crystallographic b axis [15].

At the higher magnetic field of 1.3 T [Fig. 2(c)], the magnetic response of HoFeO_3 becomes dominated by the paramagnetic polarization of the Ho^{3+} moments, especially strong along the a and b axes, consistent with previous observations [22]. As temperature decreases, the Ho^{3+} moments become increasingly polarized not only by the applied field but also via exchange coupling with the ordered Fe^{3+} sublattice. The temperature dependence of the magnetization is well fitted in this case with a Brillouin model

$$\mathbf{M}(T) = N\mu_{\text{eff}}B_J\left(\frac{\mu_{\text{eff}}\mathbf{H}_{\text{eff}}}{k_B T}\right), \quad (1)$$

where B_J is the Brillouin function for total angular momentum $\mathbf{J}$, μ_{eff} is the effective magnetic moment, and N is the number density of magnetic ions [23]. The effective magnetic field is given by $\mathbf{H}_{\text{eff}} = \mathbf{H}_{\text{app}} + \lambda\mathbf{M}$, where λ is the molecular

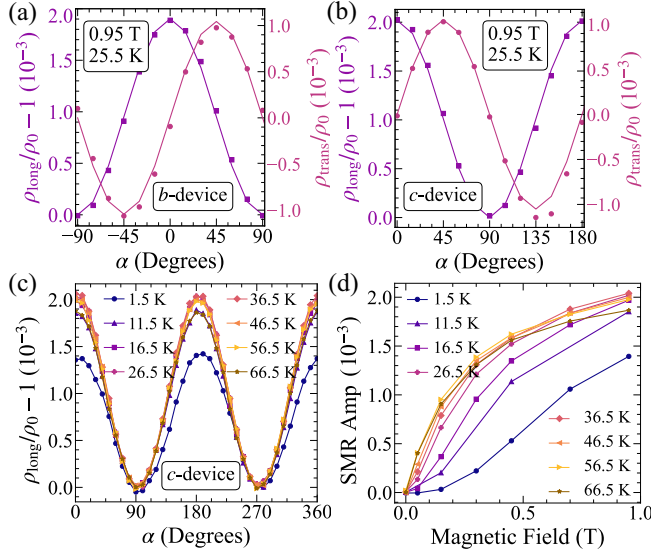


FIG. 3. In-plane angle-dependent SMR measurements for *b* and *c* devices. (a), (b) Longitudinal and transverse SMR curves measured in the *b* device (a) and *c* device (b) at 25.5 K by rotating a magnetic field of 0.95 T in the plane of the Hall bars. (c) Temperature dependence of the SMR measured in the *c* device from 1.5 K–66.5 K. (d) Magnetic field dependence of the SMR amplitude for selected temperatures.

field constant accounting for exchange coupling between Ho and Fe sublattices at lower temperatures. The model captures the gradual build up of Ho magnetization below 50 K and remains valid to the lowest temperatures. From the fit with Eq. (1), we extract $\mu_{\text{eff}} \approx 12.9 \mu_B$ along both axes. This slightly exceeds the free-ion value of Ho^{3+} ($10.61 \mu_B$), likely due to the polarization of the Fe^{3+} sublattice ($5.92 \mu_B$) via Fe-Ho exchange coupling. As discussed by Jensen and Mackintosh [24], such mean-field models incorporate collective effects—including exchange and crystal field interactions—into an effective parameter, which can enhance the fitted moment beyond the free-ion value. In the higher temperature range above 50 K, the magnetization is more accurately fitted with a Curie-like power law, $\mathbf{M} \propto T^{-\gamma}$, with fitting exponent $\gamma \approx 1.21$, close to the theoretical value for 3D Ising-like systems ($\gamma \approx 1.24$) [23].

We now proceed to the discussion of the SMR data. Figure 3 shows both longitudinal and transverse components of the SMR measured for two crystal orientations of the bar by rotating a magnetic field of 0.95 T in the plane of the device. Positive SMR signal is observed at all measured temperatures, including above the magnetic phase transition at 58 K. While the emergence of positive-SMR at low temperatures could be attributed to the magnetic contribution from the Ho sublattice, its persistence at higher temperatures in the Γ_4 phase rather connects it to the weak ferromagnetic component $\mathbf{m}_{\text{Fe}}$. This implies that the spin current in the Pt layer interacts more effectively with $\mathbf{m}_{\text{Fe}}$ than with $\mathbf{l}_{\text{Fe}}$, resulting in a ferromagneticlike SMR response even at elevated temperatures

$$\rho_{\text{long}} = \rho_0 + \rho_1 [1 - m_t^2] = \rho_0 + \frac{\rho_1}{2} (1 + \cos 2\alpha), \quad (2)$$

$$\rho_{\text{trans}} = \rho_1 m_j m_t = \frac{\rho_1}{2} \sin 2\alpha. \quad (3)$$

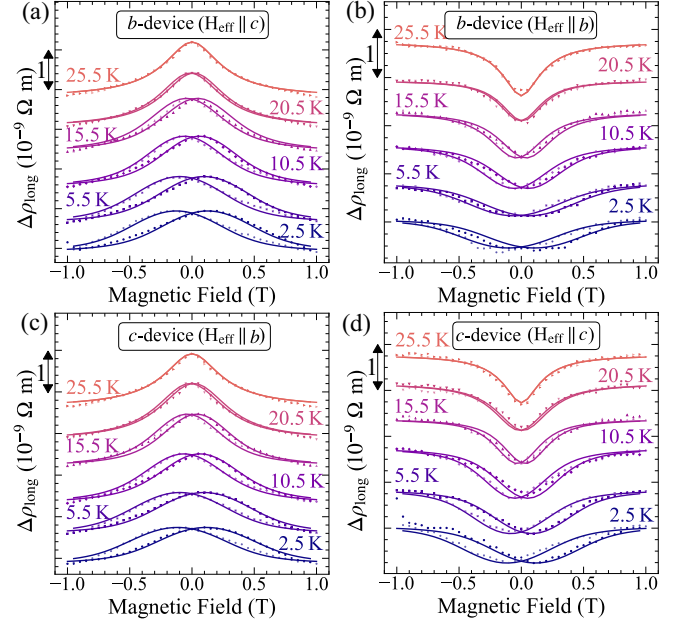


FIG. 4. Field-dependent SMR measured from -1 T to $+1$ T for both *b* and *c* devices, with the magnetic field applied along either the *b* or *c* axis in the sample plane. (a), (b) FDSMR curves for the *b* device with field along the *c* axis (a) and *b* axis (b). (c), (d) The *c*-device with field along the *b* axis (c) and *c* axis (d). In all cases, the longitudinal resistivity exhibits hysteresis below ~ 20 K, attributed to Fe-Ho exchange coupling. All temperature-dependent curves were fitted using the effective free energy model [Eq. (4)], and markers denote experimental data. For clarity, vertical offsets have been applied to curves at each temperature.

Here, ρ_0 is the angle-independent interface resistivity, ρ_1 is the SMR coefficient, m_j and m_t are the projections of $\mathbf{m}_{\text{Fe}}$ in the direction parallel and perpendicular to the current direction [25], while α is the in-plane angle between the charge current and the magnetic field, as shown in Fig. 1(b).

Different from [26], where a continuous increase in SMR amplitude with field strength was observed, in our measurements, the SMR amplitude saturates at approximately 1 T in the higher temperature range. This behavior confirms that above the spin reorientation temperature (~ 58 K), the weak ferromagnetic component $\mathbf{m}_{\text{Fe}}$, which lies in the rotation plane of the applied field, aligns parallel to the external field. The saturation thus reflects a complete alignment of the $\mathbf{m}_{\text{Fe}}$ with the external field and negligible field-induced canting. As temperature is decreased, the SMR curves are no longer saturated, suggesting an increase in anisotropy. Moreover, at the lowest temperature, the curves noticeably deviate from the fitting with Eqs. (2) and (3), which is understood with an additional in-plane anisotropy term.

In Fig. 4 we plot the magnetic field dependence of ρ_{long} for the two device orientations. Below 20 K, a hysteresis develops and grows with decreasing temperature, consistent with the onset of an in-plane anisotropy term at low temperature. In the broader sweep range shown in Fig. 5(a), a peak indicated by a red arrow appears at higher fields, and gradually shifts to lower fields as temperature is decreased. The position of this peak at 2.5 K coincides with the saturating field of the Ho^{3+}

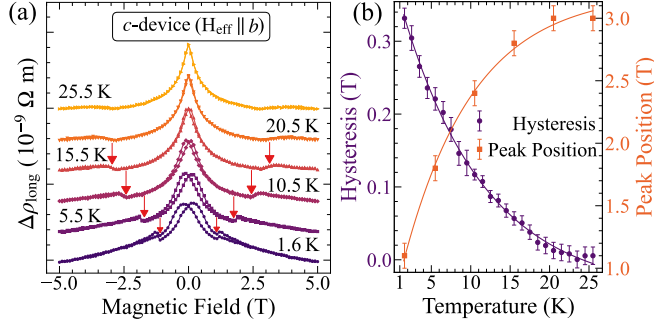


FIG. 5. Field sweep measurements of the SMR for the *c* device with in-plane magnetic field along the *b* axis. (a) Longitudinal resistivity measured from -5 T to $+5$ T at various temperatures. (b) Temperature dependence of hysteresis amplitude and field position of the peak indicated by the red arrow in (a). Fittings with exponential decay functions are obtained in each case. A vertical offset has been applied for each curve in (a) for visual clarity.

moments measured via SQUID [Fig. 2(d)]. As the temperature is decreased, stronger Fe-Ho exchange results in lower saturation field values, which explains the temperature dependence of the peak position. Simultaneously, the amplitude of the hysteresis grows [Fig. 5(b)], connecting the additional in-plane anisotropy term to Fe-Ho exchange.

The temperature dependence of both hysteresis amplitude and peak position in Fig. 5(b) were modeled using an exponential function, $H(T) \propto \exp^{\mp(T/T_0)}$, where the minus (plus) sign describes the decay (increase) of hysteresis amplitude (peak field position) with increasing temperature. Here, T_0 defines the characteristic temperature scale associated with the thermal evolution of each quantity. The nearly identical values of $T_0 \approx 10$ K obtained from both fits suggest a shared microscopic origin, likely governed by the thermal activation and exchange-mediated response of Ho^{3+} moments. The exponential suppression of hysteresis reflects the weakening of the Ho-induced pinning at elevated temperatures, while the decrease in the peak field position enhanced polarizability and alignment of Ho^{3+} moments at low temperatures.

Finally, we present the SSE data measured in the same device bars. Figure 6 shows in-plane angle rotation measurements for the *c* device at 1.6 K for various magnetic field strengths. The observed SSE voltage exhibits a clear $\sin \alpha$ dependence, typical of ferromagnets or canted AFMs, where the spin polarization of the thermally induced spin-currents follows the direction of the external magnetic field [27]. The SSE amplitude becomes significant only at very low temperatures below 4 K. This suggests that the magnetic ordering of the Ho sublattice, with the onset of a Ho-Ho exchange term, is determinant for establishing significant spin transfer along the temperature gradient. Similarly to the Fe spins, the Ho spins are expected to develop canting because of the strong positive Fe-Ho exchange interaction. It is therefore reasonable to conclude that low-energy q-FM modes of the Ho sublattice are mainly responsible for the SSE measured at low temperature.

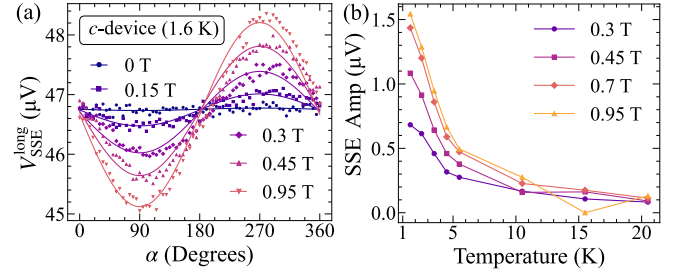


FIG. 6. In-plane SSE measurements for the *c* device, showing angular and temperature dependence of spin current generation in the low-temperature regime. (a) Angle-dependent longitudinal SSE voltage at 1.6 K under various magnetic fields, showing a clear $\sin \alpha$ dependence, consistent with ferromagneticlike spin polarization aligned along the magnetic field direction. The modulation amplitude increases with field strength, indicating enhanced spin current generation in the ordered phase. (b) SSE amplitude as a function of temperature for different magnetic fields, vanishing above 20 K due to the loss of spin polarization of the Ho sublattice at higher temperatures.

IV. DISCUSSION

The rich physics of HoFeO_3 is the result of complex $3d-4f$ interactions between the Fe and Ho sublattices. The $2J + 1 = 17$ states of Ho^{3+} are split by the crystal field into a series of singlet pairs, with the ground pair often referred to as a pseudodoublet. The energy position of the doublet centroid and its splitting are affected by Fe-Ho exchange, which increases at lower temperatures [11,28]. This induces a change in the thermal occupation of the Ho orbitals, with direct impact on the magnetic anisotropy of the Fe sublattice. The close link between the population of Ho orbitals and Fe anisotropy has been directly exploited in another orthoferrite material to excite coherent spin oscillations by impulsively modifying the magnetic anisotropy when electronic orbital transitions were forced via resonant terahertz pumping [2]. In our experiment, it is the thermal occupation of the Ho orbitals that changes, resulting in a temperature-dependent variation in the Fe anisotropy.

The SMR, SSE, and SQUID magnetometry studies collectively reveal a rich evolution of magnetic anisotropy in HoFeO_3 , driven by the temperature-dependent polarization of Ho^{3+} moments and their exchange coupling to the Fe sublattice. Based on these measurements, we identify three distinct temperature regions below the spin reorientation transition, each characterized by unique field-dependent signatures.

In the first temperature region, from 20 K to 52 K, the magnetic susceptibility of HoFeO_3 undergoes a steep increase with decreasing temperature when the magnetic field is applied along *a* and *b* axes, orthogonal to the Néel vector direction. This is the sign of higher magnetic polarizability of Ho that results from higher Fe-Ho exchange. The increasing Fe-Ho exchange interaction is also picked up in the SMR measurements in Fig. 3. Here, the SMR amplitude increases steadily in the considered temperature range. This gradual increase may reflect enhanced canting, and thus a higher value of $\mathbf{m}_{\text{Fe}}$.

The second temperature region that we identify is between 5 K to 20 K. In this region, we more evidently see the impact of increasing Fe-Ho exchange on the magnetic anisotropy of the Fe spin lattice. The SQUID measurements at 0.13 T [Fig. 2(b)] show a drop in magnetic susceptibility along the a axis below 20 K, accompanied by an increase in magnetic susceptibility along the c axis. This can be explained by the partial rotation of the easy axis away from the a -crystal direction toward the bc plane. A similar tilting was deduced in the magnetostriction measurements reported in Ref. [11] and attributed to the anisotropic energy shift of the Ho^{3+} ground pseudodoublet levels.

The SMR measurements also reflect stronger anisotropies below 20 K, accompanied by a tilting of the easy axis. Below 20 K, the 0.95 T field applied in the field rotation measurements in Fig. 3(c) is no longer high enough to saturate $\mathbf{m}_{\text{Fe}}$ in the plane of the device, explaining the observed nonsaturating trend in SMR amplitude [Fig. 3(d)]. The tilting of the easy axis introduces anisotropy in the plane of the device, responsible for the skewing of the SMR curves at low temperature and the appearance of the hysteresis in the field sweeps in Figs. 4 and 5.

The hysteresis curves are fitted for different temperatures with an effective macrospin free energy density model:

$$F = -\mu_0 \mathbf{H}_{\text{app}} \cdot \mathbf{m}_{\text{Fe}} - \mu_0 \mathbf{H}_{\text{exc}} \cdot \mathbf{m}_{\text{Fe}} - K_a m_{\text{Fe},a}^2 - K_b m_{\text{Fe},b}^2 - K_c m_{\text{Fe},c}^2. \quad (4)$$

K_a , K_b , and K_c represent the anisotropy constants along the orthorhombic crystal axes, and $m_{\text{Fe},i}$ is the projection of the Fe magnetization on the i axis. The applied field $\mathbf{H}_{\text{app}}$ couples to the Fe macrospin via the Zeeman energy, while $\mathbf{H}_{\text{exc}}$ models the exchange interaction with the Ho moments, which is taken to be in plane. In the fittings shown in Fig. 4, the exchange field direction was allowed to vary with temperature, a good fit was obtained assuming a rotation of $\mathbf{H}_{\text{exc}}$ toward the b axis as temperature was decreased.

Finally, the third temperature region is below 5 K, the temperature at which the Ho spontaneously orders. In this region, we don't observe a noticeable impact on magnetic anisotropy; instead, the SSE is strongly affected and undergoes a steep increase. The amplitude of the SSE is directly related to the low-energy magnon states, which become thermally populated in the presence of an interface temperature gradient, thereby inducing a spin current. The temperature dependence of the q-FM and q-AF modes of the Fe lattice was detected in the sub-THz range in Raman spectroscopy [29], Faraday rotation [30,31], and time-domain THz spectroscopy [32]. The q-FM frequency was observed to increase at temperatures below 40 K, in agreement with the gradual increase of anisotropy induced by the polarization of the rare earth. The increase in the SSE signal at low temperatures is instead associated with the appearance of additional lower frequency modes with the onset of Ho-Ho exchange. We note that these

modes will have a predominant orbital character and will contribute with high spin currents because of the high total angular momentum of Ho. Temperature-dependent neutron inelastic scattering studies of the spin dynamics in HoFeO_3 combined with simulations based on linear-spin wave theory allowed mapping the spin wave spectrum of the Fe sublattice and predicted the modes of the Ho sublattice to sit in the 0–1 meV energy range [14].

V. CONCLUSIONS

By combining SQUID magnetometry with spin Hall magnetoresistance (SMR) and spin Seebeck effect (SSE) measurements, we have provided a comprehensive picture of how Ho^{3+} moments interact with the Fe sublattice in HoFeO_3 and govern its low-temperature magnetic behavior. Our results reveal that the gradual polarization of Ho^{3+} ions below the spin reorientation transition significantly alters the magnetic anisotropy of the Fe spins, leading to a reorientation of the easy axis toward the bc plane. This manifests in SMR as a nontrivial field dependence and low-temperature hysteresis. At even lower temperatures, below 4 K, the onset of Ho magnetic order gives rise to a sharp increase in SSE, which we attribute to the emergence of low-energy magnon modes associated with Ho–Ho exchange. These findings establish HoFeO_3 as a model system for exploring electrically accessible low-energy spin dynamics in complex magnetic oxides, with implications for cryogenic spintronics and the engineering of spin-based devices based on 4f orbital physics.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [33], embargo periods may apply.

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