

Switching magnetic spin states using small magnetic fields in compositionally complex $\text{Sm}(\text{Ti}, \text{Cr}, \text{Mn}, \text{Fe}, \text{Co}, \text{Ni}, \text{Cu})\text{O}_3$

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High-entropy perovskites offer a unique platform for exploring magnetic phenomena arising from extreme *B*-site chemical disorder. In $\text{Sm}(M7)\text{O}_3$, where there are seven cations in equal amounts at the *B* site ($M = \text{Ti}, \text{Cr}, \text{Mn}, \text{Fe}, \text{Co}, \text{Ni}, \text{Cu}$), we observe long-range antiferromagnetic ordering near 105 K accompanied by a small but robust excess magnetic moment intrinsic to the chemically disordered lattice. This uncompensated moment is evident from zero-field-cooled – field-cooled irreversibility, shifts in the isothermal $M(H)$ loops, and discrete remanent states identified through direct-current–demagnetization measurements. Remarkably, cooling fields as small as ± 20 Oe are sufficient to select the direction of the excess moment, and the chosen magnetic state remains stable against applied fields up to 50 kOe. A low-temperature anomaly in the remanent magnetization further reveals a secondary contribution from the Sm^{3+} sublattice, although the primary origin of the excess moment resides in the *B*-site antiferromagnetic sublattice.

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I. INTRODUCTION

High-entropy oxides (HEOs) represent an emerging class of materials [1–3] in which multiple principal elements share crystallographic sites, introducing a high degree of configurational disorder while retaining long-range structural order [4–6]. This chemical complexity acts as an additional tuning parameter for magnetic interactions, often leading to behavior that departs from conventional ordered oxides. In perovskite HEOs, a recurring theme is the coexistence of long-range magnetic order with signatures of frustration, spin canting, or glassy dynamics, reflecting the broad distribution of local exchange interactions created by disorder [2,7–11]. Studies on HEO systems show that such disorder can stabilize antiferromagnetic correlations alongside spin-glass-like freezing or weak ferromagnetic contributions, indicating that competing interactions are intrinsic to these systems rather than extrinsic perturbations.

Importantly, the magnetic response in HEOs is governed by a complex interplay between lattice distortions, cation distribution, and exchange pathways, which can vary significantly

depending on whether disorder is introduced on the *A* site, *B* site, or both. As highlighted in recent reviews, compositional complexity introduces local structural inhomogeneity, exchange disorder, and competing magnetic energy scales, thereby enabling emergent phenomena such as noncollinear order, enhanced anisotropy, and metastable magnetic states [3]. This perspective establishes HEOs as model systems for exploring how disorder and competing interactions shape magnetic ground states in complex oxides.

The interplay of chemical disorder, lattice geometry, and competing exchange interactions brings forth a wide range of ground states across different structural families. In high-entropy rare-earth pyrochlores, antiferromagnetic (AFM) correlations develop but do not evolve into long-range order; instead, the system exhibits a spin-glass ground state characterized by frequency-dependent spin freezing and unsaturated magnetization, arising from geometric frustration combined with random occupation of the rare-earth sublattice [12]. When configurational disorder is confined to the *A* site, the transition-metal–oxygen–transition-metal superexchange framework remains largely preserved, and the system retains a canted antiferromagnetic ground state; however, local structural distortions and heterogeneous rare-earth–transition-metal interactions promote magnetic noncollinearity, spin-reorientation behavior, and enhanced anisotropy [13].

In several perovskite HEOs, long-range antiferromagnetic or ferrimagnetic-like order survives despite the random arrangement of multiple transition-metal cations, while the underlying disorder frequently generates ill-compensated antiferromagnetism with small residual moments embedded in an AFM lattice [7–9]. This behavior mirrors phenomena long known in classical dilute antiferromagnets such as FeF_2 and $\text{Fe}_x\text{Zn}_{1-x}\text{F}_2$, where random-field and random-exchange effects lead to uncompensated local moments, excess

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remanence, and apparent exchange-bias-like shifts even in the absence of ferromagnetic interfaces [14–16]. The FeF_2 literature demonstrates that such uncompensated moments can be stabilized by local anisotropy and disorder, giving rise to magnetic remanent states and field-history-dependent magnetization [17]. Similar signatures were recently reported in compositionally complex perovskites, including the high-entropy perovskite family, where AFM order coexists with small, frozen-in moments arising from nanoscale exchange imbalance [5].

Recent advances further show that these disorder-borne moments can be manipulated with surprisingly small cooling fields. In chemically complex fluorides and oxides, cooling fields of only a few tens of oersteds can reorient pinned AFM spin states, induce exchange-bias-like offsets, and generate large low-field magnetic switching effects [18]. In high-entropy perovskites (HEPs), such sensitivity is particularly notable because the disordered B -site network creates a rich landscape of magnetic spin configurations, as highlighted in the recent literature on magnetically ordered HEPs [5,7]. In this work, we investigate the high-entropy perovskite $\text{Sm}(M7)\text{O}_3$, where equal fractions of seven transition-metal ions populate the B site. We show that the system develops an antiferromagnetic order with a small but robust excess magnetic moment intrinsic to the high-entropy lattice. Most notably, this excess moment can be reversibly switched using cooling fields as small as ± 20 Oe, and at low temperatures, the selected magnetic state remains stable up to high applied fields. These results demonstrate that high-entropy perovskites offer a powerful route for low-field tuning of uncompensated antiferromagnetic states and suggest that such behavior may be a general feature of magnetically ordered HEOs independent of the rare-earth ion occupying the A site.

II. EXPERIMENTAL METHODS

$\text{SmTi}_x\text{Cr}_x\text{Mn}_x\text{Fe}_x\text{Co}_x\text{Ni}_x\text{Cu}_x\text{O}_3$, denoted $\text{Sm}(M7)\text{O}_3$ in the following, where $x = 1/7$, was synthesized using conventional solid-state synthesis with a final sintering temperature of 1150°C for 48 h in air. Sm_2O_3 (Alfa Aesar, >99.9%) was predried at 975°C overnight prior to weighing and combined in stoichiometric proportions with the metal oxides TiO_2 (Merck), Cr_2O_3 (Alfa Aesar), MnO_2 (Cerac), Fe_2O_3 (Alfa Aesar), Co_3O_4 , NiO (Sigma Aldrich), and CuO (Cerac), all with purity >99.5%. Co (Höganäs, >99%) was oxidized to Co_3O_4 at 600°C for 6 h, and its purity was confirmed by powder x-ray diffraction prior to synthesis. The reagents were ground with minimum (approximately 2 mL) ethanol for 30 min and pressed into a pellet. The pellet was heated in air using an alumina crucible at 900°C for 12 h, 1000°C for 24 h, 1100°C for 48 h, and 1150°C for 48 h with intermediate grindings. A heating rate of $5^\circ\text{C}/\text{min}$ and a cooling rate of $10^\circ\text{C}/\text{min}$ were used during sintering. Powder x-ray diffraction patterns were collected at room temperature using a Bruker D8 ADVANCE diffractometer equipped with the TWIN/TWIN setup (40 kV, 40 mA) using $\text{Cu K}\alpha$ radiation in Bragg-Brentano geometry. Data were collected using a Lynx-eye XE-T position-sensitive detector, operated over an angular 2θ range of 10° – 80° with a step size of 0.02° corresponding to a Q range of 0.75 – $4.95 \text{ \AA}^{-1}$. Rietveld re-

finements of the structural model were completed using the FullProf program [19]. The sample composition was obtained using a ZEISS Leo 1550 field emission scanning electron microscope equipped with an AZtec energy dispersive X-ray detector for spectroscopy analysis (EDS). Pellets of the sintered powder were attached to conducting carbon tape, and point EDS measurements were performed using an accelerating voltage of 20 kV on nine spots (see Fig. SM1 in the Supplemental Material) [20]. The magnetic properties, including the temperature-dependent zero-field-cooled (ZFC) and field-cooled (FC) magnetization, and ac susceptibility were acquired using a superconducting quantum interference design (SQUID) measurement system from Quantum Design Inc. Magnetic field (H) dependent magnetization (M) measurements were recorded on the same equipment at a constant temperature. $M(H)$ hysteresis curves were recorded at different temperatures. The direct current demagnetization (DCD) was recorded by cooling the sample in a large magnetic field (-50 kOe) and applying and removing a reverse (positive) magnetic field of increasing amplitude [21]. The remanence value after each reverse field pulse is plotted as a function of the reverse field in DCD plots.

III. RESULTS AND DISCUSSION

Figure 1 presents the powder X-ray diffraction pattern of the high-entropy perovskite $\text{Sm}(M7)\text{O}_3$, together with the corresponding Rietveld refinement. The results show that the compound crystallizes with a distorted perovskite structure, adopting orthorhombic $Pnma$ symmetry [$a = 5.5797(7) \text{ \AA}$, $b = 7.6515(7) \text{ \AA}$, $c = 5.3778(5) \text{ \AA}$]. The calculated profile provides a good fit to the observed data aside from one very weak reflection present at $2.49 \text{ \AA}^{-1}$ which arises from trace amounts of an unknown impurity. The high quality of the fit and observed phase purity suggest that the Sm has been fully incorporated into the A site of the perovskite. The transition metals are randomly distributed over the octahedral B sites, and the Sm cations are located on the 12-fold oxygen coordination A sites. The result of EDS point analysis of nine spots indicates an average cation composition of $\text{Sm}_{51(1)}\text{Ti}_{9.7(9)}\text{Cr}_{7.4(5)}\text{Mn}_{7(1)}\text{Fe}_{7.5(5)}\text{Co}_{7.2(5)}\text{Ni}_{5(1)}\text{Cu}_{3.0(5)}$. This is close to the expected ratio for the B -site metals of 7.14. On average, the sample is deficient in Cu, most likely as a result of the low melting point of CuO . The structural model highlights the extensive chemical disorder on the B site, where seven distinct transition-metal cations occupy the octahedral framework in a completely random manner. This high configurational complexity—characteristic of high-entropy oxides—is expected to produce significant local distortions and competing exchange pathways, which play a central role in the magnetic behavior discussed in later sections. Figure 2(a) shows the temperature dependence of the magnetization measured under an applied field of $H_{\text{DC}} = 50$ Oe following ZFC and FC protocols, after resetting the magnet to ensure a near-zero cooling field. Both the ZFC and FC curves exhibit a clear onset of magnetic ordering near $T \approx 105$ K. Below this temperature, a pronounced bifurcation between the ZFC and FC branches is observed, with the FC magnetization remaining significantly larger than the ZFC magnetization over the entire low-temperature range.

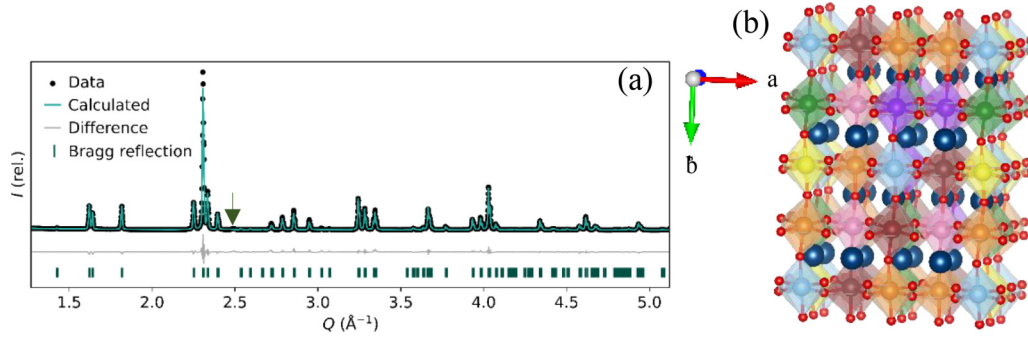


FIG. 1. (a) Rietveld refinements (green line) of powder x-ray diffraction data (black circles), with Bragg reflections (green tick marks). $R_{wp} = 7.97\%$, and $\chi^2 = 60.0$. One weak reflection arising from an unidentified impurity is identified with an arrow. (b) The crystal structure of $\text{Sm}(\text{M7})\text{O}_3$ derived from Rietveld analysis, with a randomly distributed B site (multicolored spheres and octahedra); Sm is shown by dark blue spheres, and O is shown by red spheres.

The magnetization decreases below ~ 10 K. The in-phase component of the ac susceptibility χ_{ac} , measured at frequencies of 1.7, 17, and 170 Hz, is shown in Fig. 2(b). A prominent feature is observed near $T \approx 105$ K, a sharp maximum of the magnetic transition coinciding with the magnetic transition seen in the dc magnetization data. Within the measured frequency range, the position of this feature shows negligible frequency dependence. Figure 3(a) shows the temperature dependence of the magnetization measured under an applied field of $H_{meas} = 1$ kOe after cooling the sample in small fields of $+20$ and -20 Oe. For $H_{CF} = +20$ Oe, the magnetization curve remains positive over the entire temperature range. For $H_{CF} = -20$ Oe, the magnetization is negative at low temperatures, and the two branches converge near the magnetic ordering temperature (~ 105 K) for both cooling conditions. Figure 3(b) displays isothermal field-dependent $M(H)$ magnetization curves measured under ZFC conditions at 2, 25, and 75 K. At 2 K, the magnetization varies approximately linearly with the applied magnetic field over the entire measured range up to ± 50 kOe. As the temperature increases, coercivity and

hysteresis appear. The slope of the $M(H)$ curves decreases with increasing temperature as well. In the inset, a zoomed view of the $M(H)$ curve at 2 K is shown, which, in fact, is not symmetrical across the origin and rather seems shifted up on the magnetization scale or to the negative side on the field axis. This observation, together with the positive ZFC magnetization in Fig. 2(a), suggests a small positive background field in the measurement system which plays the same role as the $+20$ Oe cooling field in Fig. 3(a). Figure 3(c) presents the temperature-dependent magnetization measured at a higher applied field of $H_{meas} = 50$ kOe (instead of 1 kOe) after cooling the sample in small opposite fields of -20 and $+20$ Oe. The magnetization curves remain similar at high temperatures, but the one cooled in -20 Oe deviates from the one cooled in $+20$ Oe below 25 K. The magnetization values at low temperatures thus depend on the sign of the cooling field. The results shown in Fig. 3(c) suggests that the magnetic susceptibility of the material includes three contributions, χ_{Sm} , the paramagnetic susceptibility of the Sm^{3+} sublattice; χ_{3d} , which represents the field- and temperature-dependent

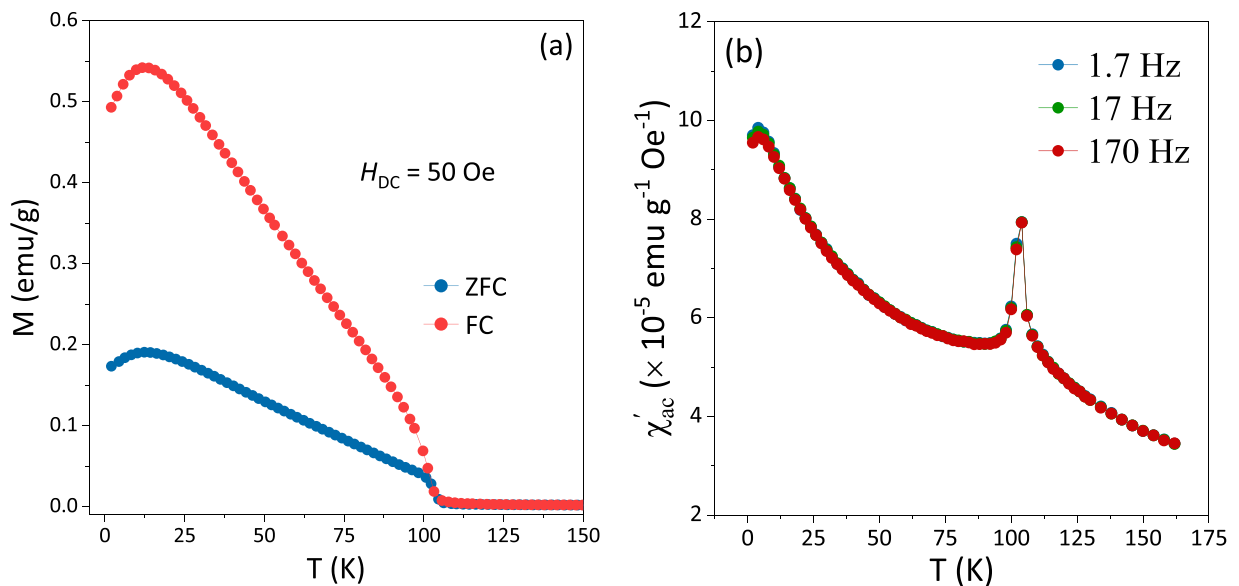


FIG. 2. (a) Temperature-dependent magnetization in an applied field of $H_{DC} = 50$ Oe under ZFC and FC protocols after the magnet is reset to ensure the cooling field is near zero. (b) In-phase component of ac susceptibility (ac amplitude $h_{ac} = 4$ Oe).

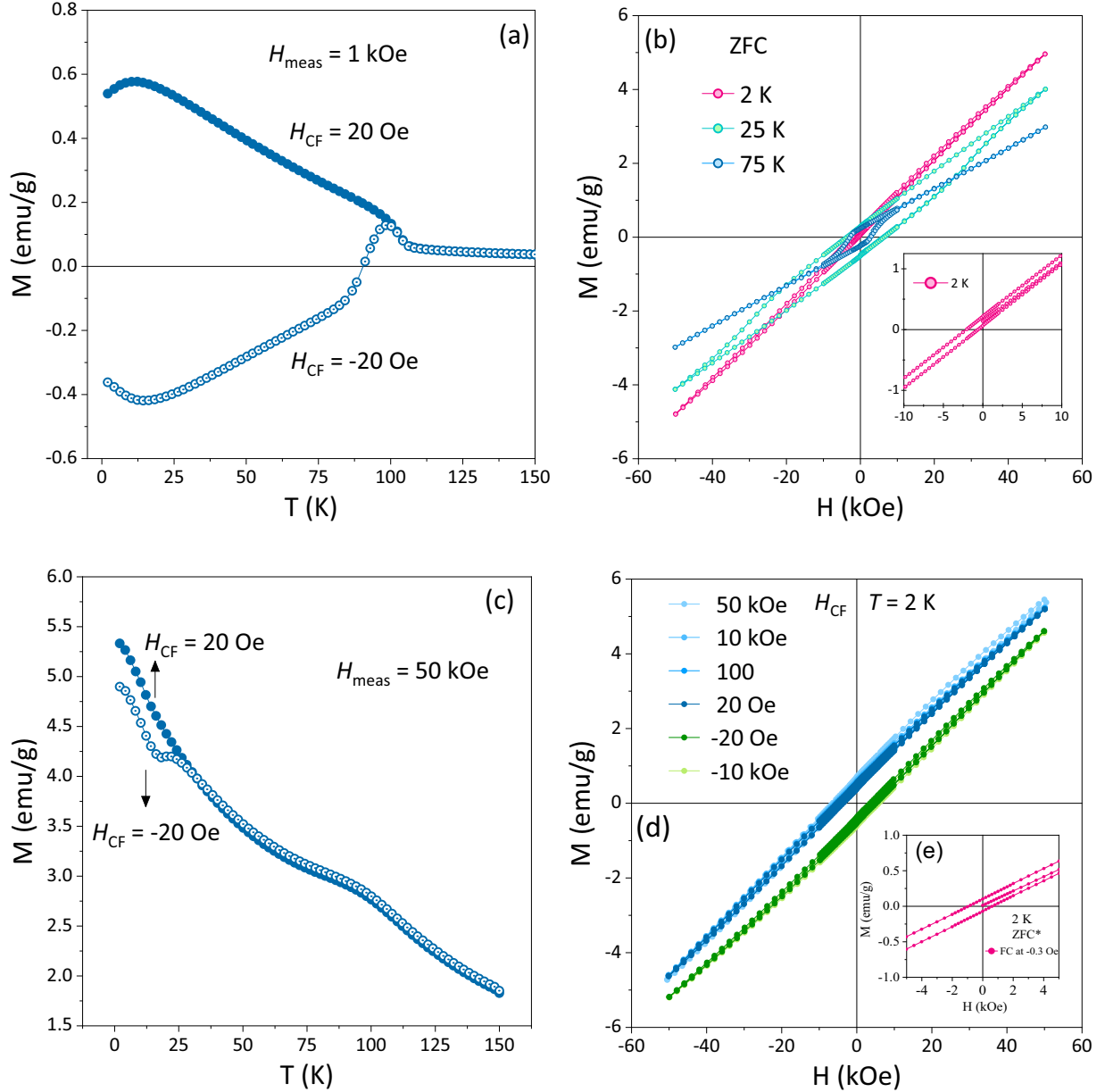


FIG. 3. Magnetization curve recorded versus temperature after cooling in a small field of -20 or 20 Oe: (a) $H_{\text{meas}} = 1$ kOe (see Fig. SM4 for complete ZFC/FC curves) and (c) $H_{\text{meas}} = 50$ kOe [see Fig. SM5(a) for complete ZFC/FC curves] [20]. (b) Field-dependent magnetization curves at 2, 25, and 75 K. The inset shows the zoomed version of the ZFC $M(H)$ loop at 2 K. (d) Magnetization curves recorded versus field after cooling in the same fields as well as fields of larger magnitude (see legend: positive and negative cooling fields are plotted using shades of blue and green, respectively). (e) $M(H)$ recorded under the ZFC* condition, where ZFC* (FC at -0.3 Oe) indicates that a small background correction was made to obtain a field as close as possible to zero field.

susceptibility of the B -site cations responsible for the AFM order near ~ 105 K; and M_0 , an excess moment arising from the ill-compensated sublattices of the AFM structure, as

$$M(H, T)/H = \chi_{\text{Sm}}(H, T) + \chi_{3d}(H, T) + M_0(H, T)/H.$$

The direction of the excess moments is determined by the sign of the cooling field. Figure 3(d) presents field-dependent magnetization curves measured at 2 K under FC conditions for several cooling fields ranging from -10 to $+50$ kOe. All $M(H)$ curves exhibit approximately linear field dependence up to ± 50 kOe. The slopes of the curves are similar for different cooling fields, but the curves are vertically or horizontally

shifted relative to each other depending on the magnitude and sign of the cooling field. Interestingly, the shift depends relatively little on the strength of the field but is highly sensitive to the sign of the cooling field. Figure 3(e) shows a magnified view of the low-field region of the $M(H)$ curves at 2 K. A small hysteresis is visible, and the measurements taken under ZFC* conditions (which is field cooling at -0.3 Oe to compensate for the background field and achieve a near-zero effective cooling field as confirmed by series of FC measurements; see Fig. SM3 [20]) closely follow the central branch, indicating minimal offset compared to curves obtained after cooling in larger positive or negative fields. $M(H)$ at higher temperatures

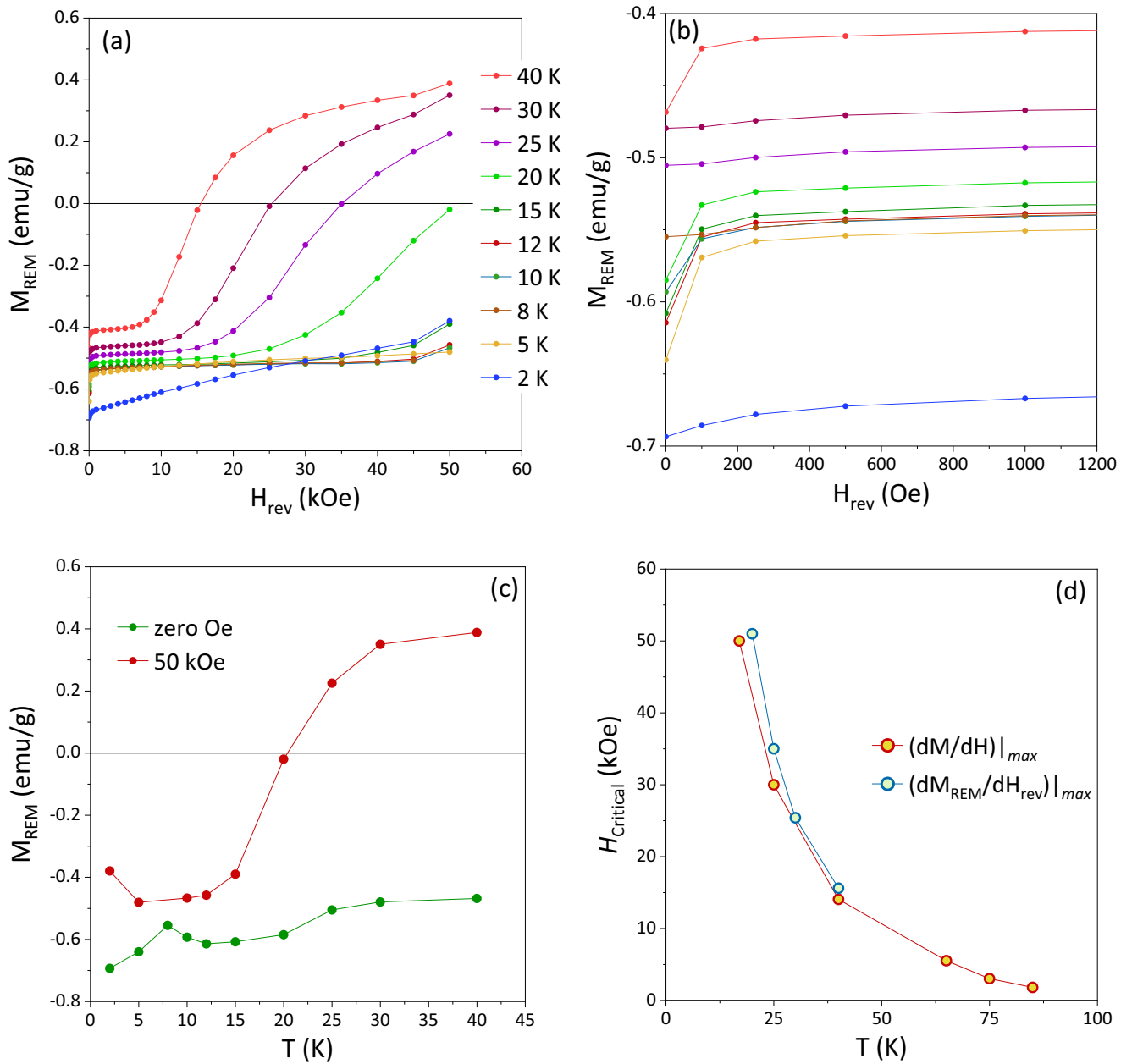


FIG. 4. (a) DCD remanence curves obtained after cooling in -50 kOe and applying and removing a reverse magnetic field of increasing magnitude from 0 to $+50$ kOe. (b) Zoomed view of (a). (c) The values of the remanence at 0 Oe and after a 50 kOe field pulse are plotted with respect to temperature. (d) Temperature dependence of H_{Critical} extracted from $(dM/dH)_{\text{max}}$: the maximum slope in $M(H)$ curves shown in Fig. SM6 and $(dM_{\text{REM}}/dH_{\text{rev}})_{\text{max}}$ identified as the switching field for which M_{REM} is zero (i.e., switches from negative to positive) in (a).

[as shown in Fig. 3(b) and Fig. SM6 [20]] shows a significant coercivity, which is not in reach at low temperature. It is noteworthy that ΔM (which equals M after cooling in $+20$ Oe minus M after cooling in -20 Oe) at 50 kOe measured from $M(H)$ curves and ΔM from the $M(T)$ curves after cooling in $+20$ and -20 Oe and measured under 50 kOe are the same.

To probe the switching of the magnetization after field cooling, we performed a series of DCD measurements. DCD has extensively been used to probe the magnetization switching of magnetic materials and is used here to investigate the stability of the magnetic configurations under increasing magnetic field. In this protocol, the sample is cooled from

the paramagnetic state to the target temperature in a strong negative field (-50 kOe), the field is removed at 2 K, and the magnetization is subsequently recorded after applying and removing reverse fields until $+50$ kOe is reached for each temperature. The remanent magnetization M_{REM} after each reverse field pulse is recorded and plotted as a function of the reverse field in Fig. 4. The resulting $M_{\text{REM}}(H_{\text{rev}})$ curves, shown in Figs. 4(a) and 4(b), display a characteristic two-step evolution at all temperatures between 2 and 40 K. The magnetization initially remains on a low-moment plateau [Fig. 4(b)] and then undergoes an abrupt increase at a well-defined switching field [Fig. 4(a)]. Notably, the magnitude of this

switching field coincides with the critical fields $H_{\text{Critical}}(T)$ extracted from the maxima in dM/dH obtained under ZFC conditions, as shown in Fig. 4(d). Coercivity appears in the $M(H)$ loops [Figs. 3(b) and SM6] because of the hysteretic behavior of the excess moment $M_0(H, T)$. Defining the coercive field as the field required to flip the excess moment, this coercive field always increases with decreasing temperature and reaches field strengths larger than the maximum field of the experiments (5 T) at temperatures below 40 K. The coercivity field extracted directly from the hysteresis curves is measurable for all curves above 25 K, but the curves measured at lower temperatures do not reach high enough fields to open the hysteresis curves and thus do not provide coercive fields. The temperature dependence of the remanent magnetization $M_{\text{REM}}(T)$, plotted in Fig. 4(c), provides additional insight because the zero-field values of M_{REM} exhibit a pronounced anomaly below ~ 10 K which suggests the Sm contribution to the magnetic signal [see the $M(T)$ curves at low T in Fig. 2].

The present results demonstrate that the antiferromagnetic order established in $\text{Sm}(M7)\text{O}_3$ near 105 K is accompanied by a small but robust excess magnetic moment. This uncompensated moment manifests through ZFC-FC irreversibility, vertical shifts of the $M(H)$ curves after field cooling, and discrete remanent states observed in DCD remanence measurements. This behavior is inconsistent with a fully compensated collinear antiferromagnet [22] and instead indicates intrinsic ill-compensation within the ordered AFM structure. Given the statistically distributed multicomponent B site, this excess moment is most naturally attributed to local imbalance of AFM sublattices in each grain of the polycrystalline sample arising from chemical disorder and competing exchange interactions inherent in high-entropy perovskites. A salient feature of this uncompensated moment is its strong sensitivity to the cooling field. Remarkably, cooling fields as small as ± 20 Oe are sufficient to select the sign of the magnetization, and the selected state remains stable even under applied fields as large as 50 kOe. This behavior indicates that the excess moment is associated with magnetic configurations separated by sizable anisotropy barriers, such that a weak symmetry-breaking field can bias the system into one of two energetically equivalent but oppositely oriented states [23]. Once selected, these states exhibit high stability, pointing to an effective anisotropy that pins the uncompensated component of the antiferromagnetic structure. At low temperatures, an additional contribution from the Sm^{3+} sublattice becomes evident. The anomaly observed below ~ 10 K in the remanent magnetization, in this temperature range, suggests that the Sm moments couple antiferromagnetically to the excess moments. However, the primary origin of the excess moment and its field tunability resides in the B -site-driven AFM framework rather than in the rare-earth (RE) contribution. Importantly, these observations are not expected to be unique to Sm-based compounds. The emergence of a controllable uncompensated moment appears to be a generic consequence of an AFM order in chemically disordered HEPs, independent of the specific RE ion occupying the A site. This finding is supported by observations from prior works which discussed the magnetic ground state of HEPs as predominantly governed by the transition-metal (B -site) sublattice, while A -site disorder

primarily plays a secondary structural role. This distinction is particularly evident when comparing A -site- and B -site-disordered perovskites.

In A -site-disordered rare-earth perovskites, such as $(\text{Gd}_{0.2}\text{Nd}_{0.2}\text{La}_{0.2}\text{Sm}_{0.2}\text{Y}_{0.2})\text{CoO}_3$, the magnetization remains largely linear and nonsaturating over a wide field range, consistent with dominant paramagnetic or antiferromagnetic correlations [24]. Only a weak hysteresis is observed at low temperatures, with negligible intrinsic exchange bias (EB). The magnetic response in these systems is instead governed by spin-state transitions and competing interactions within the Co network, while A -site configurational disorder primarily modifies structural parameters such as lattice distortion and bandwidth. These observations indicate that A -site disorder alone does not generate a significant exchange imbalance or pinned uncompensated spins. A qualitatively different behavior emerges when disorder is introduced directly on the B site. In B -site-disordered high-entropy perovskites, such as $\text{La}(\text{SB}_{0.2})\text{O}_3$, the $M(H)$ loops remain unsaturated even at high fields and exhibit large coercivity, reflecting strong magnetic anisotropy and frustration [5]. Importantly, these systems display a pronounced vertical EB-like shift, which has been attributed to uncompensated spins embedded within an antiferromagnetic matrix, together with small ferromagnetic clusters arising from competing exchange pathways among different transition-metal ions. The coexistence of AFM and locally FM interactions leads to a spatially heterogeneous magnetic state, in which field cooling stabilizes pinned moments that produce loop asymmetry.

This disorder-driven picture is further supported by compositionally tunable high-entropy perovskite films, where the nature of EB evolves with the balance between antiferromagnetic and ferromagnetic interactions [25]. In the antiferromagnetic-dominated regime, vertical loop shifts are observed, consistent with isolated uncompensated moments, while increasing ferromagnetic connectivity leads to the emergence of horizontal EB associated with coupled AFM/FM regions. At still higher ferromagnetic fractions, the exchange bias disappears altogether. These results demonstrate that EB in HEPs is not an interfacial effect, but rather a bulk property controlled by the competition and percolation of exchange interactions within the disordered B -site network.

Similar conclusions have been drawn for high-entropy spinel oxides, where horizontal exchange bias arises intrinsically from exchange coupling between antiferromagnetic regions and weak ferromagnetic components stabilized by chemical disorder [26]. The absence of well-defined phase boundaries in these systems underscores that exchange bias can emerge from intrinsic magnetic heterogeneity and frustration within a single-phase material.

The microscopic origin of these effects is consistent with established models of exchange bias [27–29]. Classical theory attributes exchange bias to pinned uncompensated spins that establish unidirectional anisotropy during field cooling, while experimental studies have shown that even a small fraction of such pinned spins can produce measurable loop shifts. Importantly, it has been demonstrated that exchange-bias-like behavior can arise intrinsically in single-phase antiferromagnets, where frustration and finite-size effects generate uncompensated moments within

the bulk. In such cases, vertical loop shifts naturally emerge from pinned bulk spins rather than interfacial coupling, providing a direct analog to disorder-driven exchange bias in high-entropy oxides.

IV. CONCLUSIONS

In this work, we reported the ability to reversibly tune and stabilize distinct magnetic states using fields as small as ± 20 Oe in a compositionally complex perovskite with seven B -site cations, $\text{Sm}(M7)\text{O}_3$. We speculate that this is an inherent feature of HEPs, owing to the intrinsically ill-compensated AFM structure in those systems. These results thus indicate that this low-field tuning may be a generic and technologically relevant feature of magnetically ordered HEPs.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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