

Characterization of the magnetic phase transitions in double perovskite $\text{Nd}_2\text{NiMnO}_6$

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Through detailed analysis of neutron powder diffraction data, we confirm that the double perovskite $\text{Nd}_2\text{NiMnO}_6$ adopts a monoclinic $P2_1/n$ structure with nearly complete B -site ordering of Ni^{2+} and Mn^{4+} cations. Below $T_1 = 198$ K, magnetic susceptibility measurements and further analysis of neutron diffraction data reveal that the Ni^{2+} and Mn^{4+} sublattices undergo ferromagnetic ordering, as expected of strong $3d$ - $3d$ exchange interactions. Upon cooling through $T_2 = 22$ K, a secondary magnetic transition is observed, below which we have discovered an additional noncollinear symmetry-breaking order of Nd^{3+} moments. We argue that the canting of the rare-earth moments naturally arises through the competition of f - d and f - f Heisenberg exchange interactions, which may be finely balanced in the double perovskite framework. Furthermore, the symmetry of the ground state magnetic structure implies significant Nd^{3+} easy plane anisotropy, and an effective decoupling of the antiferromagnetic spin canting from the transition metal sublattice. Between T_1 and T_2 , $\text{Nd}_2\text{NiMnO}_6$ shows anomalous behavior in the frequency-dependent ac magnetic susceptibility that is characteristic of reentrant spin-glass-like properties attributed to antisite disorder and competing interactions. Finally, analysis of isothermal magnetization yields magnetic entropy changes that suggest $\text{Nd}_2\text{NiMnO}_6$ and related compounds have potential for magnetic refrigeration, showing a peak in the magnetic entropy change of $2.25 \text{ J kg}^{-1} \text{ K}^{-1}$ at T_1 under a 7 T field. The scaling behavior of the magnetic entropy, paired with analysis of other critical exponents, shows that the ferromagnetic transition at T_1 is consistent with a mean-field second-order phase transition. Taken together, our results provide crucial details on the magnetic properties of $\text{Nd}_2\text{NiMnO}_6$ and further consolidate the expectation that double perovskites may serve as model systems for investigating competing magnetic interactions, magnetocaloric effects, and reentrant spin-glass behavior.

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I. INTRODUCTION

The ABO_3 perovskite oxides are one of the most versatile families of crystal structures and have been duly explored throughout the study of complex magnetic materials [1]. The versatility emerges primarily due to patterns of BO_6 octahedral tilts and rotations [2], typically described using Glazer notation [1,3,4], that can accommodate size mismatches among A and B cations and oxygen anions as quantified by the Goldschmidt tolerance factor. A vast range of perovskite materials have hence been synthesized with magnetic cations at both the A and B sites. Derived from this structural framework are the rare-earth (R) double perovskites (DPs), with general chemical formula $R_2\text{BB}'\text{O}_6$. Here, three distinct crystallographic sites are now capable of accommodating different

magnetic ion species, where the B and B' sites are usually occupied by transition metal ions [5–7].

In the presence of typical $a^-a^-c^+$ octahedral tilting, disorder among, say, B^{3+} and B'^{3+} cations maintains orthorhombic $Pnma$ symmetry. However, rock-salt ordering of, say, B^{2+} and B'^{4+} cations driven by electrostatic forces lowers the crystal symmetry to monoclinic $P2_1/n$. Notably, many such ordered DP compounds exhibit ferromagnetism (FM), which arises due to exchange interactions between the B^{2+} and B'^{4+} ions [8]. This scenario is distinct from the ternary ABO_3 parent compounds which are typically antiferromagnetic (AFM). In some DPs, antisite disorder (ASD) of B and B' cations can introduce vastly more complex magnetic states [9,10]. For example, in the nominally ordered DP $\text{La}_2\text{NiMnO}_6$, both FM and AFM orders coexist. The FM coupling is attributed to $\text{Ni}^{2+}(\text{t}_{2g}^6\text{e}_g^2)\text{-O}^{2-}\text{Mn}^{4+}(\text{t}_{2g}^3\text{e}_g^0)$ exchange, while AFM coupling is associated with the $\text{Ni}^{2+/3+}\text{-O-Ni}^{2+/3+}$ and $\text{Mn}^{3+/4+}\text{-O-Mn}^{3+/4+}$ exchange induced by the ASD, which together lead to spin or cluster-glass states below 70 K [11]. Unveiling the complex interplay between these exchange interactions and how they couple to local structural distortions leading to phenomena such as magnetocrystalline anisotropy and multiferroicity [12] is quintessential for designing multifunctional materials [13].

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It was recently shown that replacing La^{3+} with magnetic R^{3+} ions such as Nd^{3+} , i.e., $\text{Nd}_2\text{NiMnO}_6$, can dramatically modify the magnetostructural phase diagram. $\text{Nd}_2\text{NiMnO}_6$ crystallizes in a nominally B^{2+} and B^{4+} cation ordered monoclinic $P2_1/n$ crystal structure. We note that both computational and experimental evidence have confirmed the presence of Ni^{2+} and Mn^{4+} ions and refuted the possibility of mixed valence states providing clarity on the compound's magnetic dynamics [14,15]. Introducing Nd^{3+} into the structure results in a reduced Ni-O-Mn bond angle, weakening the ferromagnetic exchange and lowering T_1 to ~ 200 K [16,17]. A long-range ordered ferromagnetic structure has been proposed below T_1 [18]. A second magnetic transition near ~ 100 K was reported and assigned to valence state fluctuations e.g., $\text{Ni}^{3+}/\text{Mn}^{3+}$ [18] that may be attributed to ASD-induced frustration resulting in cluster-glass behavior [19]. Furthermore, interactions between the Nd^{3+} and transition metal sublattices are expected to become significant at temperatures below 50 K [17]. Indeed, this magnetic interplay has been highlighted by the low temperature ordering of the Nd moments relative to the Ni-Mn magnetic structure, as indicated by neutron diffraction studies [20].

In this article, we report a detailed study of the structural and magnetic properties of $\text{Nd}_2\text{NiMnO}_6$ using dc and ac magnetometry combined with neutron powder diffraction. Our findings confirm the presence of Ni^{2+} and Mn^{4+} cations, with no evidence of Ni^{3+} or Mn^{3+} . We show that magnetic ordering develops below $T_1 \sim 198$ K, with neutron diffraction confirming FM Ni-Mn exchange coupling [14]. Just below 100 K, anomalies in the ac magnetization suggest reentrant spin-glass behavior, which may be attributed to frustration between sublattice interactions in the presence of ASD, as evidenced by our mixed occupancy structural refinements. We have discovered a subtle kink in the ac susceptibility near $T_2 = 22$ K, consistent with a second magnetic phase transition, which occurs concomitantly with the previously reported downturn in dc magnetization at low temperature [17–19]. Analysis of neutron diffraction data measured at 1.5 K demonstrates the onset of complex, noncollinear magnetic order of Nd^{3+} ions that contrasts previous studies [14]. Here, we find that the noncollinear order naturally arises from a competition between FM coupling of Nd^{3+} and transition metal ions (f - d exchange), and AFM coupling of Nd^{3+} ions (f - f exchange). Further, we note that spin-orbit coupling within the Nd 4*f* manifold and weak Dzyaloshinskii-Moriya interactions may contribute additional complexity in the presence of noncollinear spin arrangements. Finally, critical behavior analysis and magnetocaloric studies underscore the multifunctionality of $\text{Nd}_2\text{NiMnO}_6$, reinforcing its potential in magnetic applications.

II. EXPERIMENT

A polycrystalline sample of $\text{Nd}_2\text{NiMnO}_6$ was synthesized by a modified sol-gel route, using glycine ($\text{C}_2\text{H}_5\text{NO}_2$) as a chelating agent. In a sequential process, 2 g of $\text{C}_2\text{H}_5\text{NO}_2$ was slowly added to a 2 M starting solution containing a stoichiometric amount of $\text{Nd}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, and $\text{Mn}(\text{CH}_2\text{CO}_2)_2 \cdot 4\text{H}_2\text{O}$, with high purity (Aldrich, 99.97%) in a proportion of 2:1:1 (Nd:Ni:Mn). The prepared

sample solutions were kept at 200 °C for 24 h on a hot plate for evaporation and xerogel formation. Subsequently, all the swollen xerogels were ground and precalcined at 1000 °C for 12 h to remove all organic solvent substrates. The resulting powder was successively pulverized and pressed at 80 MPa to form a dense pellet. The final calcination was carried out at 1200 °C for 24 h to help obtain the pure DP phase. The sample's formation and phase purity were initially analyzed at room temperature by x-ray diffraction using a Rigaku DMAX Ultima+ diffractometer with monochromatic Cu- $K\alpha$ radiation, operating at 40 kV and 40 mA. Data were collected over a 2θ range of 5° to 90° with a step size of 0.02° in 2θ and a counting time of 20 s per step. The instrument was configured with a fixed divergence slit of 1/4°, an antiscattering slit of 1/2°, and a sample mask of 10 mm. dc magnetometry measurements were conducted using a superconducting quantum interference device (SQUID) magnetometer (MPMS-3, Quantum Design) within the temperature range of 2–300 K, with a maximum applied magnetic field (H) of ± 70 kOe. Demagnetization protocols were employed to ensure minimal bias from any trapped flux within the superconducting magnet. ac magnetometry measurements were performed using a Quantum Design Physical Properties Measurement System at ISIS Neutron and Muon Source, UK, with excitation fields of 0.5, 1, and 2 Oe, on cooling in zero dc field from 300 to 2 K. The measurement was repeated for 11, 110, 320, and 540 Hz excitation frequencies.

Neutron powder diffraction (NPD) measurements were conducted using the WISH diffractometer [21] at ISIS Neutron and Muon Source, UK. A 1.5485 g sample was loaded into a 6-mm-diameter vanadium can, and mounted within a ^4He cryostat. High-counting-statistics measurements, each lasting 60 min, were conducted in the paramagnetic (PM) phase at 230 K and in the magnetic phases at 100 K (high temperature) and 1.5 K (low temperature). Subsequent crystal and magnetic structure refinements were performed using the FULLPROF suite [22]. Symmetry analysis was performed using the ISOTROPY software suite [23], including the ISODISTORT tool [24].

III. RESULTS AND DISCUSSION

A. Crystal structure

The cation ordered monoclinic ($P2_1/n$) DP crystal structure is shown in Fig. 1, where Nd atoms occupy general positions (4e), while Ni and Mn are distributed across two symmetry inequivalent sublattices (2a and 2b, respectively). Three symmetry inequivalent oxygen sublattices (O1, O2, and O3) are located on general positions (4e). This structural model was refined against NPD data measured within the paramagnetic phase (at 230 K). Excellent agreement between the model and data were achieved [see Fig. 2(a)], and the refined structural parameters are summarized in Table I, which include lattice parameters, atomic positions, isotropic thermal factors, bond valence sum (BVS), selected bond distances and bond angles, and refinement reliability factors. We note that the absence of discernible impurity peaks up to the limits of instrumental sensitivity confirmed that our $\text{Nd}_2\text{NiMnO}_6$ sample was phase-pure. The degree

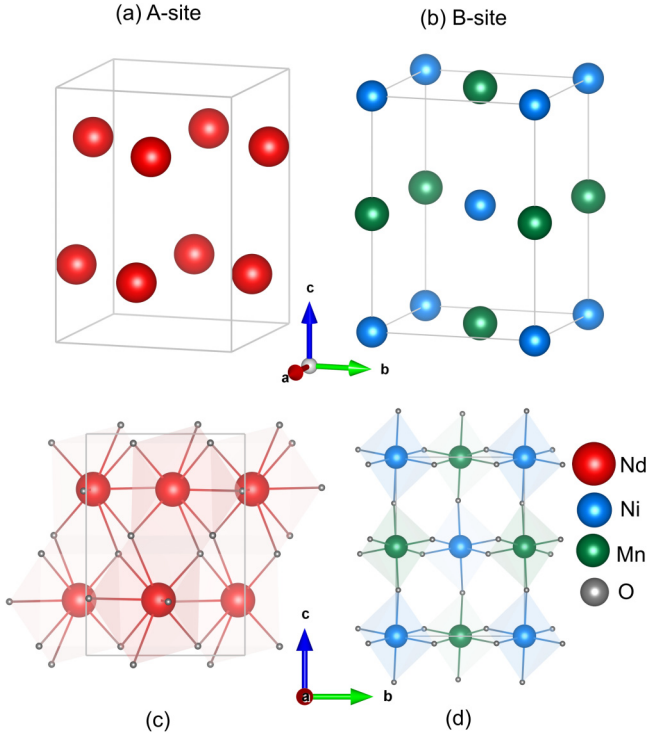


FIG. 1. The cation ordered crystal structure of $\text{Nd}_2\text{NiMnO}_6$ (monoclinic $P2_1/n$) presented as a single unit cell. (a) The A site with Nd atoms is depicted in red. (b) The B and B' sites are shown, with Ni and Mn atoms represented in blue and green, respectively. (c), (d) Projections of the crystal structure along the a axis illustrating the $(\text{Nd})\text{O}_8$ cuboctahedral coordinations and the $(\text{Ni}, \text{Mn})\text{O}_6$ tilted octahedral coordinations, respectively. The oxygen anions are represented by grey spheres.

of long-range cation ordering between Ni and Mn sites was refined against the NPD data, revealing 9% ASD (Table I). The refined lattice parameters $a = 5.40779(1) \text{ \AA}$, $b = 5.47668(1) \text{ \AA}$, $c = 7.66482(2) \text{ \AA}$, with $\beta = 90.02769(4)^\circ$ and a unit cell volume of $V = 227.007(1) \text{ \AA}^3$ corroborates those previously published [10,11,14,20]. The BVS analysis is consistent with the expected valences of Nd^{3+} , Ni^{2+} , and Mn^{4+} . Local crystal structure analysis reveals significant bond length and angle variations, indicating a highly distorted crystalline structure within the ordered $\text{Nd}_2\text{NiMnO}_6$ system. The Ni-O bond lengths are significantly longer than the Mn-O bond lengths, consistent with well-established cation order (and the calculated BVS), and the Ni-O-Mn bond angles indicate a significant departure from 180° due to octahedral tilting, resulting in complex magnetic exchange pathways [19,25]. Using refined lattice parameters, the octahedral rotation angles θ (octahedra tilt about the pseudocubic $(110)_p$ axis), ϕ [octahedral rotation about the pseudocubic $(001)_p$ axis], Φ [combined tilt about the threefold $(111)_p$ axes], and Θ (octahedra distortion from average bonds) were calculated to quantify deviations from the ideal cubic perovskite configuration [26]. The angles were estimated as follows: $\theta = \cos^{-1}(a/b) \approx 9.10^\circ$, $\phi = \cos^{-1}(\sqrt{2}a/c) \approx 3.82^\circ$, $\Phi = \cos^{-1}(\sqrt{2}a^2/bc) = \cos^{-1}(\cos \gamma \cos \phi) \approx 9.86^\circ$, and $\Theta = [180 - \langle \text{Ni-O-Mn} \rangle]/2 \approx 12.87^\circ$.

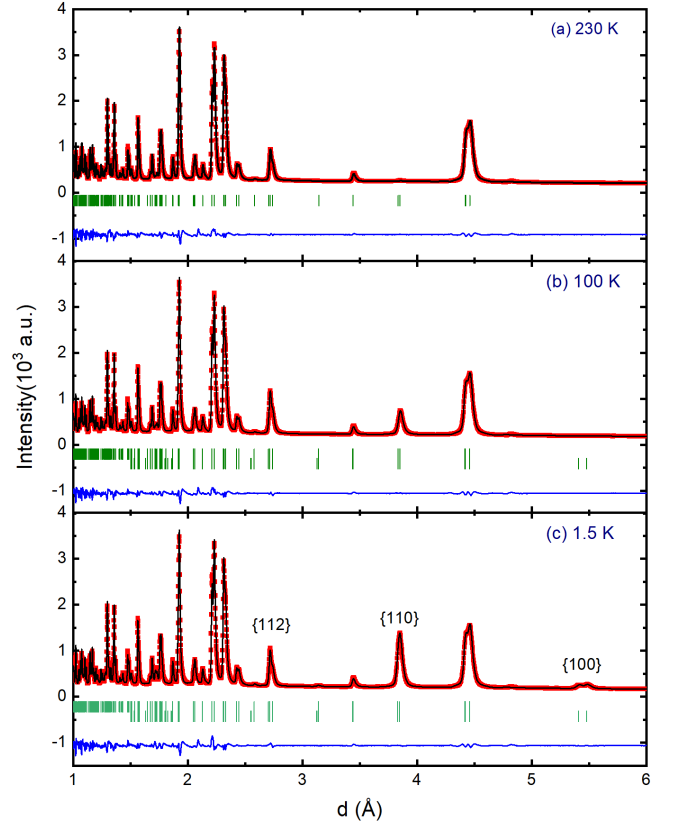


FIG. 2. Neutron powder diffraction data measured in three magnetic phases: (a) paramagnetic (230 K), (b) first ordered (100 K), and (c) ground state (1.5 K). Data are shown as red points, the fitted patterns as black lines, and the difference curve $I_{\text{obs}} - I_{\text{calc}}$ as blue lines at the bottom of the panes. The top and bottom row of green tick marks in each pane indicate the position of nuclear and magnetic Bragg peaks, respectively.

B. dc magnetometry

The dc magnetic susceptibility χ , measured as a function of temperature, is presented in Fig. 3(a). Both field-cooled-cooling (FCC) and zero-field-cooled (ZFC) data exhibit a sharp increase in susceptibility on cooling below $\approx 200 \text{ K}$, likely signaling the onset of ferromagnetic or ferrimagnetic (FiM) long-range magnetic order. On further cooling, a downturn in the susceptibility can be seen below $\approx 50 \text{ K}$. These observations are consistent with previous reports [10,11,17,25]. Below $\approx 200 \text{ K}$, the FCC and ZFC curves bifurcate, indicating a small degree of irreversibility due to spin freezing or blocking associated with magnetic anisotropy. The inset to Fig. 3(a) shows $\chi(T)$ measured at higher fields ($H = 5$ and 10 kOe), where broadening of the transition and suppression of bifurcation are evident.

Figure 3(b) shows the linear region of the inverse susceptibility χ^{-1} vs T (above T_1 in the region of $215\text{--}300 \text{ K}$), suggesting paramagnetic behavior well described by the Curie-Weiss law:

$$\chi^{-1} = \frac{T - \theta_{\text{CW}}}{C}, \quad (1)$$

where C is the Curie-Weiss constant. Fitting to this linear region gave a positive Curie-Weiss temperature of

TABLE I. Structural parameters of the $\text{Nd}_2\text{NiMnO}_6$ sample obtained through Rietveld refinement from NPD data at 230 K. The lattice parameters were determined to be $a = 5.4078(1) \text{ \AA}$, $b = 5.4767(1) \text{ \AA}$, and $c = 7.6648(2) \text{ \AA}$, with $\beta = 90.0277(4)^\circ$ and a unit cell volume of $V = 227.007(1) \text{ \AA}^3$. Bond valence sums (BVSs) were calculated using the parameters $R_0(\text{Nd}^{3+}) = 2.66$, $R_0(\text{Ni}^{2+}) = 2.17$, $R_0(\text{Mn}^{4+}) = 1.753$, and $B = 0.37$, where the bond valence $\text{BV} = \exp((R_o - R)/B)$. N.B. For the BVS, the majority cation is used for mixed occupancy sites.

Structural parameters				
Atom	Fractional coordinates	$U_{\text{iso}} (\times 10^{-2} \text{\AA}^2)$	BVS ($ e $)	Occupancy
Nd	$x = 0.0097(6)$ $y = 0.5445(3)$ $z = 0.7495(4)$	0.98(1)	+3.27	Nd
Ni	$x = y = z = 0$	1.06(1)	+1.94	91%Ni 9% Mn
Mn	$z = 0.5$	1.06(1)	+3.59	91%Mn 9% Ni
O1	$x = 0.0787(5)$ $y = 0.0167(4)$ $z = 0.2575(6)$	1.26(7)		O
O2	$x = 0.2165(5)$ $y = 0.2967(6)$ $z = -0.0420(9)$	0.42(1)		O
O3	$x = 0.2030(6)$ $y = 0.2823(6)$ $z = 0.5404(10)$	1.42(2)		O
Bond angles				
$\langle \text{O1-Ni-O1} \rangle$		180.0(0)		
$\langle \text{O1-Ni-O2} \rangle$		89.9(3), 90.1(3)		
$\langle \text{O1-Ni-O3} \rangle$		89.5(3), 90.5(3)		
$\langle \text{O1-Mn-O1} \rangle$		180.0(0)		
$\langle \text{O1-Mn-O2} \rangle$		89.3(3), 90.7(3)		
$\langle \text{O1-Mn-O3} \rangle$		89.5(3), 90.5(3)		
$\langle \text{Ni-O1-Mn} \rangle$		154.38(2)		
$\langle \text{Ni-O2-Mn} \rangle$		153.9(4)		
$\langle \text{Ni-O3-Mn} \rangle$		154.5(4)		
Bond lengths				
Ni-O1		2.021(5)		
Ni-O2		2.029(4)		
Ni-O3		2.024(4)		
Mn-O1		1.909(5)		
Mn-O2		1.922(4)		
Mn-O3		1.921(4)		
Reliability factors				
χ^2		11		
$R_{\text{P}}(\%)$		4.55		
$w_{\text{P}}(\%)$		3.04		
R_{Bragg}		3.82		

$\theta_{\text{CW}} \approx 206(2) \text{ K}$, consistent with unfrustrated and dominant FM interactions. The calculated effective magnetic moment (μ_{eff}) was found to be $4.69(1) \mu_{\text{B}}/\text{f.u.}$ which is in excellent agreement with the theoretical value of $\mu = 4.80 \mu_{\text{B}}/\text{f.u.}$ (considering spin only moments, where $g_L = 2$, $S = 1$ and $S = \frac{3}{2}$ for Ni^{2+} and Mn^{4+} , respectively), hence confirming the oxidation states obtained by BVS analysis, above. The

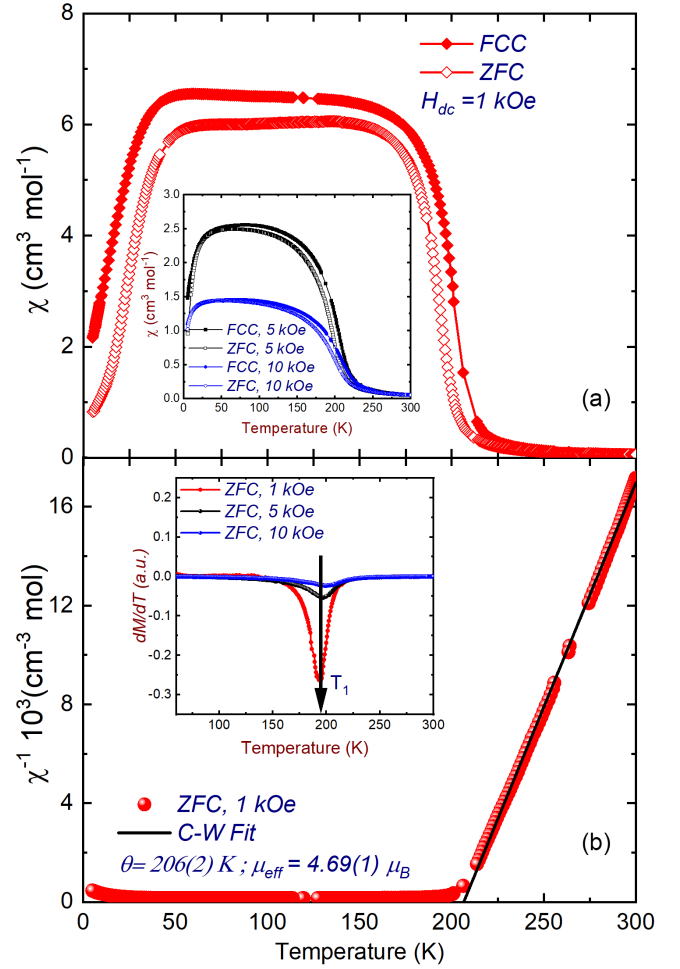


FIG. 3. (a) Temperature dependence of the dc magnetic susceptibility of $\text{Nd}_2\text{NiMnO}_6$ measured under ZFC and FCC conditions in an applied field of 1 kOe. The inset shows a comparison of M - T curves (ZFC/FCC) at different H_{dc} fields. (b) The inverse susceptibility ($\chi^{-1} = H/M$) as a function of temperature. The black line shows the paramagnetic region's best fit using the Curie-Weiss law.

inset derivative dM/dT highlights the first transition, where a pronounced dip is observed at $T_1 \approx 198 \text{ K}$, marking the onset of FM/FiM ordering. Furthermore, the field dependence in peak position ($H = 5$ and 10 kOe) is noticeable, where the position of T_1 shifts to a higher temperature with increasing field.

Figure 4 shows isothermal $M(H)$ measurements performed at various temperatures. At $T = 300 \text{ K}$, the field dependent magnetization is linear, as expected for the PM state. Upon cooling below the FM transition at T_1 , the magnetization increased with an S-like shape without any sign of hysteresis (see, e.g., data measured at $T = 100 \text{ K}$), but with remarkably rapid switching typically characteristic of a FiM or weak FM. The linear fit of the high-field region at $T = 100 \text{ K}$ (see inset of Fig. 4) yielded $M_S = 3.366(1) \mu_{\text{B}}/\text{f.u.}$, which is lower than the theoretically calculated value likely due to thermal fluctuations. Upon cooling to 2 K , the isothermal magnetization adopts a subtle double hysteresis loop, which may be indicative of frustrated interactions arising from the interplay of rare-earth and transition metal sublattices [27,28].

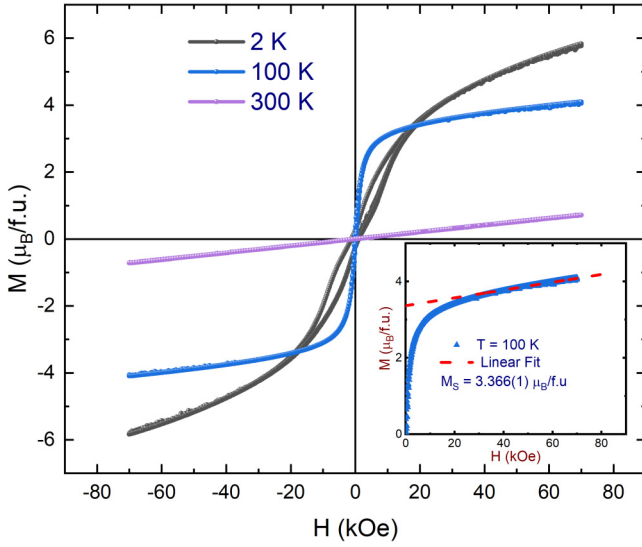


FIG. 4. Isothermal $M(H)$ curves conducted at different temperatures under applied fields up to ± 70 kOe. The inset shows the linear extrapolation of saturated magnetization at 100 K.

A complete set of isothermal magnetization measurements are given in Appendix A, which are used to show that the critical exponents that characterize phase transition at T_1 are consistent with mean-field behavior.

C. ac magnetometry

The temperature dependence of the real, $\chi'(T)$, and imaginary, $\chi''(T)$, components of the ac magnetic susceptibility is shown in Fig. 5. Both components show a sharp maximum at $T_1 \approx 200$ K, which does not shift in temperature upon changing the excitation frequency; characteristic of a phase transition to long-range magnetic order [19,29,30]. The real component is frequency independent at all temperatures and shows a drop in susceptibility below 50 K resembling the features observed in the FCC-ZFC dc magnetization (Fig. 3). To the contrary, below T_1 , $\chi''(T)$ shows a strong frequency dependence, with a broad maxima in the temperature range of 50–100 K shifting to higher temperatures as the frequency is increased. The magnitude of these shifts is consistent with reentrant glassy behavior of spin clusters reported in other materials [31–33]. True reentrance occurs when FM long-range order vanishes at lower temperatures and is replaced by a spin-glass state stabilized by disorder and competing interactions. However, our neutron powder diffraction results discussed below clearly show a large degree of long-range order is maintained down to the lowest measured temperatures. This discrepancy can be reconciled by the suggestion that at reentrance, a randomly canted ferromagnetic state may be realized at low temperature, rather than a pure spin-glass phase [33]. At low temperature, $\chi''(T)$ shows a sharp, frequency-independent anomaly at $T_2 \approx 22$ K [see inset to Fig. 5(b)], consistent with a second, low temperature magnetic phase transition. We note that $\chi'(T)$ was independent of the amplitude of the excitation field, while $\chi''(T)$ showed a small uniform increase between T_1 and T_2 on increasing the excitation from 0.5 to 2 Oe.

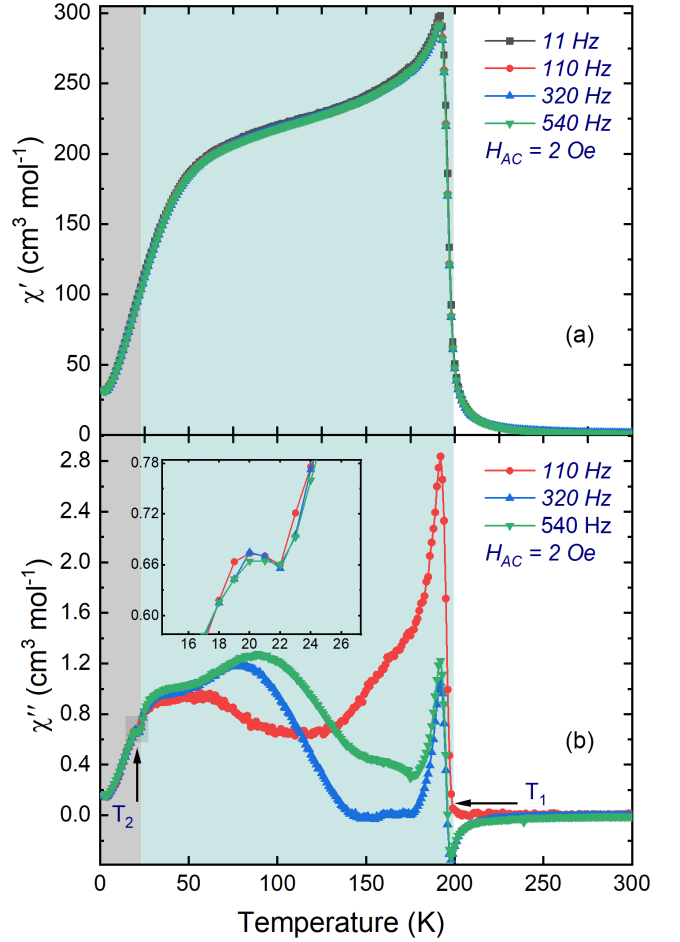


FIG. 5. The real (a) and imaginary (b) parts of the ac magnetic susceptibility of $\text{Nd}_2\text{NiMnO}_6$, measured as a function of temperature at different frequencies under $H_{dc} = 2$ Oe. The inset shows an expanded view of T_{N2} .

D. Neutron powder diffraction

Notable changes in the NPD data were observed as the sample temperature decreased, consistent with the magnetic susceptibility anomalies described above. Below $T_1 = 198$ K, magnetic peaks emerged that could be indexed as $\{112\}$, $\{110\}$, and $\{100\}$ relative to the $P2_1/n$ crystal structure [see Fig. 2(b)]. The magnetic peak widths were comparable to those of the nuclear reflections, indicating the onset of well-established long-range magnetic order. Furthermore, as the magnetic peaks coincided with the nuclear Bragg positions, the respective magnetic structure must have a Γ -point propagation vector, $\mathbf{k} = (0, 0, 0)$.

Magnetic symmetry analysis was performed using ISODISORT. Table II defines symmetry-adapted modes for each magnetic sublattice, labeled F_i , A_i , C_i , and G_i , in terms of relative signs of the magnetic moment components at each site. Linear combinations of these modes that transform by single irreducible representations are shown towards the bottom of Table II, where the polarization (spin direction) of the mode is noted in subscript relative to the monoclinic b axis. Only two irreducible representations (Γ_1^+ and Γ_2^+) enter into the decomposition of the magnetic representation for Ni and Mn sites,

TABLE II. Magnetic modes that span the four symmetry-equivalent A -site Nd ($4e$) ions and the two symmetry-equivalent B -sites ($2a$ for Ni, $2b$ for Mn) within a single $P2_1/n$ unit cell. We define four modes for the Γ -point propagation vector, $\mathbf{k} = (0, 0, 0)$: F_i , A_i , C_i , and G_i along with their components, which describe the relative alignment of magnetic moments across the symmetry-related atomic sites. The subscript i represents the directional component of the magnetic moment relative to the monoclinic b axis.

	Fractional coordinates			Magnetic modes			
A Site	x	y	z	F_i	A_i	C_i	G_i
Nd1	0	$\frac{1}{2}$	$\frac{3}{4}$	+	+	+	+
Nd2	$\frac{1}{2}$	0	$\frac{3}{4}$	+	−	−	+
Nd3	0	$\frac{1}{2}$	$\frac{1}{4}$	+	+	−	−
Nd4	$\frac{1}{2}$	0	$\frac{1}{4}$	+	−	+	−
B site							
Ni1	0	0	0	+	+		
Ni2	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	+	−		
Mn1	0	0	$\frac{1}{2}$	+	+		
Mn2	$\frac{1}{2}$	$\frac{1}{2}$	0	+	−		
Irrep	Nd			Ni		Mn	
Γ_1^+	$F_{\parallel}, A_{\perp}$			$F_{\parallel}, A_{\perp}$		$F_{\parallel}, A_{\perp}$	
Γ_2^+	$A_{\parallel}, F_{\perp}$			$A_{\parallel}, F_{\perp}$		$A_{\parallel}, F_{\perp}$	
Γ_1^-	$G_{\parallel}, C_{\perp}$						
Γ_2^-	$C_{\parallel}, G_{\perp}$						

while four irreps ($\Gamma_1^{\pm}$ and $\Gamma_2^{\pm}$) appear in the decomposition for Nd. We focus first on Γ_1^+ and Γ_2^+ magnetic structure solutions under the robust assumption that the magnetic transition metal ions play a central role in the stability of long-range magnetic order. Symmetry adapted modes of the former symmetry (Γ_1^+) involve a FM F mode aligned parallel to the monoclinic b axis, supplemented by an AFM A -type mode perpendicular to b . Modes of the latter symmetry (Γ_2^+) are similar, but with the F mode perpendicular to the monoclinic b axis and an A -type mode parallel to b . These magnetic structures correspond to the magnetic space groups $P2_1/n.1(14.75)$ and $P2_1'/n'(14.79)$, respectively.

Two magnetic structure models that included magnetic moments on the A , B , and B' sites, and transforming by Γ_1^+ and Γ_2^+ , respectively, were tested against the NPD data measured at $T = 100$ K. Rietveld refinements achieved a reasonably good fit for both models, however, detailed examination conclusively identified that the Γ_2^+ magnetic structure model (space group $P2_1'/n'$ (No. 14.79, basis = $[1, 0, 0]$, $[0, 1, 0]$, $[0, 0, 1]$, origin = $[0, 0, 0]$) was found to reproduce the observed magnetic intensities precisely. The initial, fully general model included nine free mode amplitudes, but we could readily exclude Nd moments at $T = 100$ K, which refined to values close to zero within their standard uncertainty. The absence of Nd magnetic moments at this temperature is consistent with the typical behavior of A -site rare-earth ions in perovskite manganites, which are known to order at lower temperatures due to weak f - d interactions [34]. Subsequent refinements also excluded the B and B' site A -type mode amplitude, as the fit to the data was insensi-

TABLE III. Magnetic structure parameters refined at 100 K and 1.5 K. The moment directions are defined in spherical coordinates [35,36] $m_x = m \cos(\phi) \sin(\theta) \|\mathbf{a}$, and $m_y = m \sin(\phi) \sin(\theta) \|\mathbf{b}$ and $m_z = m \cos \theta \|\mathbf{c}^*$, such that $\phi = 0$ constrains moments to the ac -plane. In this case, $\mathbf{m} = [m_x, 0, m_z]$ represents the polarization of magnetic modes $F_{\perp}$, and $A_{\perp}$.

	100 K	1.5 K	
	$m\Gamma_2^+ (F_{\perp})$	$m\Gamma_1^+ (A_{\perp})$	$m\Gamma_2^+ (F_{\perp})$
Ni			
$m (\mu_B)$	1.75(1)		1.89(1)
$\theta (^{\circ})$	143.56(2)		103.08(2)
Mn			
$m (\mu_B)$	2.18(1)		2.45(1)
$\theta (^{\circ})$	143.56(2)		103.08(2)
Nd			
$m (\mu_B)$		$\pm 0.68(4)$	$-0.91(2)$
$\theta (^{\circ})$		193.08(2)	103.08(2)
R (%)	4.56		4.87
wR (%)	3.98		4.42
R_{mag} (%)	1.47		2.29

tive to this subtle staggered magnetization along the b axis, hence the F modes were constrained to be collinear within the ac plane. The final refinement yielded excellent agreement factors ($R = 4.56\%$, $wR = 3.98\%$, $R_{\text{mag}} = 1.47\%$) with just three free parameters, confirming the reliability of the model. Magnetic moments for $1.75(1) \mu_B/\text{Ni}^{2+}$ and $2.18(1) \mu_B/\text{Mn}^{4+}$, with a polar angle $\theta = 143.56(2)^{\circ}$ were found (as detailed in Sec. III). The refinement result is shown in Fig. 2(b), and the magnetic structure is depicted in Fig. 6(a), which is fully consistent with dominant d - d exchange interactions between Ni^{2+} and Mn^{4+} ions.

At 1.5 K, additional magnetic reflections ($\{100\}$ and $\{010\}$) appeared in the neutron diffraction data, concomitant with a redistribution of intensity in the magnetic peaks discussed above. These observations are consistent with a low temperature transition at $T_2 = 22$ K (see Fig. 5) to a ground-state magnetic structure that is distinct from that refined at 100 K. The magnetic structure maintains the Γ -point propagation vector ($\mathbf{k} = (0, 0, 0)$) in the ground state, and returning to our general Γ_2^+ magnetic structure model, we found that the observed changes in the NPD data could be partly accounted for by a rotation of the Ni and Mn FM moments within the ac plane, plus an additional $F_{\perp}$ mode located on the Nd sublattice (Γ_2^+), but with moments aligned antiparallel relative to the Ni and Mn sublattices, clearly indicating that AFM f - d exchange interactions become significant at low temperature.

A complete description of the 1.5 K NPD data required an additional Nd $A_{\perp}$ mode that transforms under Γ_1^+ , resulting in a non-collinear magnetic structure with moments constrained to the ac plane. The admixture of Γ_1^+ and Γ_2^+ modes lowers the symmetry of this structure to $P\bar{1}.1$ (No. 2.4), with a basis defined by $[1, 0, 0]$, $[0, 1, 0]$, $[0, 0, 1]$ and origin at $[0, 0, 0]$. The final refinement yielded excellent agreement between observed and calculated profiles with factors ($R = 4.87\%$, $wR = 4.42\%$, $R_{\text{mag}} = 2.29\%$), confirming the reliability of the model. For the $F_{\perp}$ modes, magnetic moments of

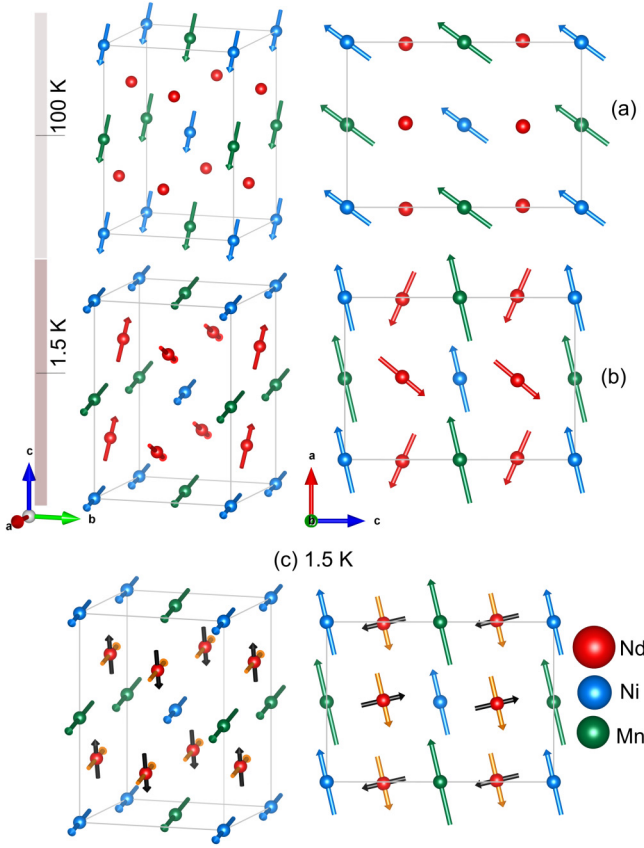


FIG. 6. Refined magnetic structures of $\text{Nd}_2\text{NiMnO}_6$: (a) $m\Gamma_2^+$ structure at 100 K, (b) $m\Gamma_1^+ + m\Gamma_2^+$ structure at 1.5 K, and (c) decomposition of $F_\perp$ and $A_\perp$ Nd moment components within the ac plane. Nd, Ni, and Mn atoms are represented in red, blue, and green, respectively, with the crystallographic unit cell outlined in grey.

$1.89(1) \mu_B/\text{Ni}^{2+}$, $2.45(1) \mu_B/\text{Mn}^{4+}$ and $-0.91(2) \mu_B/\text{Nd}^{3+}$, with a polar angle $\theta = 103.08(2)^\circ$, were found. The additional $A_\perp$ mode is associated with an Nd magnetic moment component of $0.68(4) \mu_B$ with a polar spin angle of $\theta = 193.03(2)^\circ$ in the ac plane (constrained to be perpendicular to the FM Nd moment component such that (i) symmetry equivalent Nd ions carry the same saturated moment in the ground state, and (ii) the model refinement converged to a robust solution). The refinement result is shown in Fig. 2(c), and the magnetic structure is depicted in Figs. 6(b) and 6(c). These findings contradict the previously reported ground state magnetic structure [20]. However, the above noncollinear ordering of Nd moments can be naturally understood in terms of competing antiferromagnetic f - d and f - f exchange interactions. The former introduces antiparallel alignment of the Nd F mode [red arrows in Fig. 6(c)] with respect to the transition metal moments, while the latter introduces an orthogonal, superposed Nd A mode [black arrows in Fig. 6(c)]. At the mean-field level, the canting angle will then depend on the relative strength of the f - d and f - f exchange. We note that the Nd A mode must break symmetry to lie within the ac plane, as opposed to alignment along the b axis allowed within the Γ_2^+ magnetic symmetry. This symmetry breaking may be

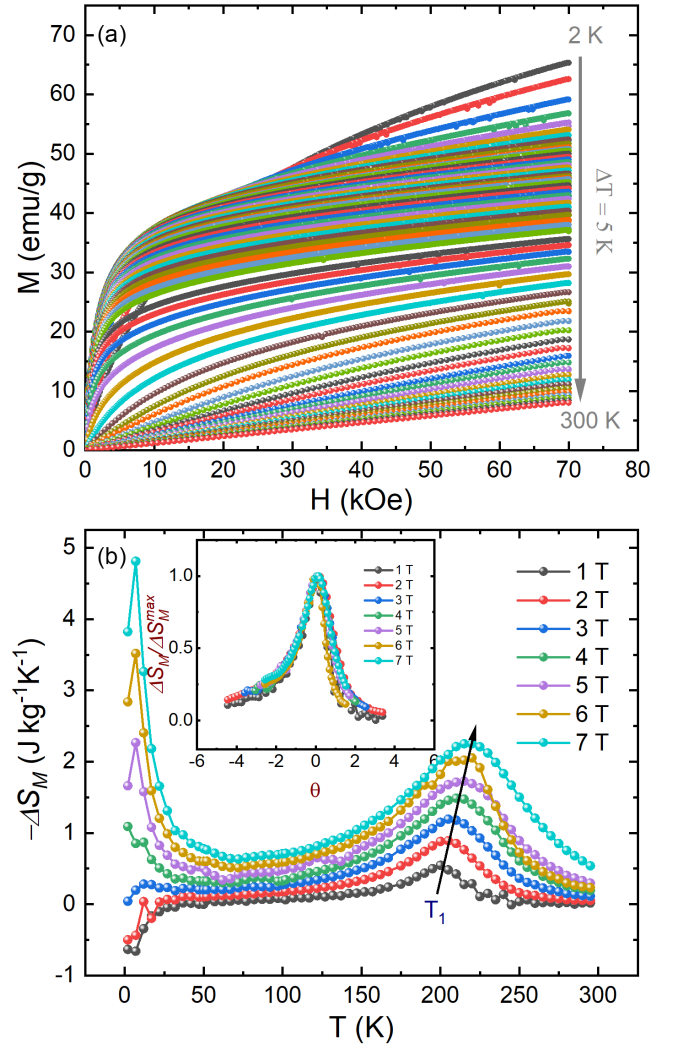


FIG. 7. (a) Isothermal 1st quadrant $M(H)$ curves conducted at $T = 2 - 300$ K with an interval of $\Delta T = 5$ K. (b) Thermal profile of field-induced magnetic entropy change, $-\Delta S_M$, estimated from isothermal magnetization curves from $T = 2 - 300$ K, under various applied magnetic fields. Inset shows the plot of normalized ($\frac{\Delta S_M}{\Delta S_M^{\max}}$) versus reduced temperature θ .

driven by significant easy-plane anisotropy of the Nd^{3+} ions and, interestingly, effectively decouples the Nd A mode from the transition metal ferromagnetic structure.

E. Magnetocaloric effect

The net change in magnetic entropy ($-\Delta S_M$) can be quantitatively estimated using isothermal magnetization $M(H)$ data, providing insight into the magnetocaloric effect (MCE) and associated magnetic transitions. As shown in Fig. 7(a), the $M(H)$ curves measured over the field range of 0 – 7 T at temperatures from 2 – 300 K demonstrate a sharp increase at low fields, followed by a linear rise without approaching saturation at high fields. The finite change in total magnetic entropy of a system due to a change in temperature (ΔT) and

magnetic field (ΔH) is expressed as

$$\begin{aligned}\Delta S_{MT} &= S_M(H + \Delta H, T + \Delta T) - S_M(H, T) \\ &= \int_H^{H+\Delta H} \left[\frac{\partial M(H, T + \Delta T)}{\partial T} \right]_H dH \\ &\quad + \int_T^{T+\Delta T} \frac{C'_H(H, T)}{T} dT.\end{aligned}\quad (2)$$

To aid analysis of discrete magnetization data the above equation can be approximated as [37,38]

$$\Delta S_M(T) = \frac{(M(T) - M(T + \Delta T))}{\Delta T} H, \quad (3)$$

where $M(T)$ and $M(T + \Delta T)$ represent values of the isothermal magnetization measured at temperatures T and $T + \Delta T$, at an applied field of H . Note that in this case we have taken the change in field ΔH to be relative to zero [39]. Figure 7(b) shows the temperature dependence of $-\Delta S_M$ for different fields. ΔS_M exhibits a broad peak centered at T_1 decreasing symmetrically on both sides as expected for a second-order FM-PM phase transition, and below $T < 50$ K anomalous behavior occurs likely due to the ordering of Nd^{3+} rare-earth ions [8].

The peak in $-\Delta S_M(T)$ at T_1 increases with increasing field, and shifts to higher temperatures. At $H = 7$ T, a maximum entropy change of $-\Delta S_M^{\max} = 2.25 \text{ J kg}^{-1} \text{ K}^{-1}$ is observed, which is smaller than some values reported for Ni/Mn-based double perovskites [15,40] but somewhat larger than previous measurements on the Nd system [8] (see Appendix B for a comparative summary). We note that $-\Delta S_M^{\max}$ follows a power law dependence on field, $-\Delta S_M^{\max} \propto H^n$, with $n = 0.667(1)$ as shown in Fig. 8(a). This value agrees strongly with the mean-field ferromagnetic value of $n = 2/3$ [41–43].

The MCE at T_1 was further examined using a normalizing universal scaling law. First, values of $-\Delta S_M(T)$ for a given applied field were scaled by a factor $\Delta S_M / \Delta S_M^{\max}$, and, second, the temperatures were scaled to a reduced temperature $\theta_{\pm}$ defined as follows [42]:

$$\theta_- = \frac{T_{\max} - T}{T_{\text{cold}} - T_{\max}}, \quad T < T_{\max}, \quad (4)$$

$$\theta_+ = \frac{T - T_{\max}}{T_{\text{hot}} - T_{\max}}, \quad T > T_{\max}, \quad (5)$$

where $T_{\max}$ denotes the temperature at $-\Delta S_M^{\max}$, and T_{cold} and T_{hot} are the temperatures of two reference points (below and above T_1) at full width half maximum of the peak in $-\Delta S_M$. As highlighted in the inset of Fig. 7(b), the rescaled curves collapse onto a single unified curve; behavior typical of a second-order FM phase transition.

The relative cooling power (RCP) parameter is typically used to evaluate the cooling efficiency of candidate MCE materials. The RCP can be determined from the $-\Delta S_M(T)$ curves using the equation [44,45]

$$\text{RCP} = |\Delta S_M^{\max}| \Delta T_{\text{FWHM}}, \quad (6)$$

where $\Delta T_{\text{FWHM}} = T_{\text{hot}} - T_{\text{cold}}$. The RCP value as a function of the magnetic field H is displayed in Fig. 8(b), which shows a monotonic increase with ΔH from $\sim 13 \text{ J kg}^{-1} \text{ K}^{-1}$ at 1 T to $\sim 157 \text{ J kg}^{-1} \text{ K}^{-1}$ at 7 T. The field dependence could be

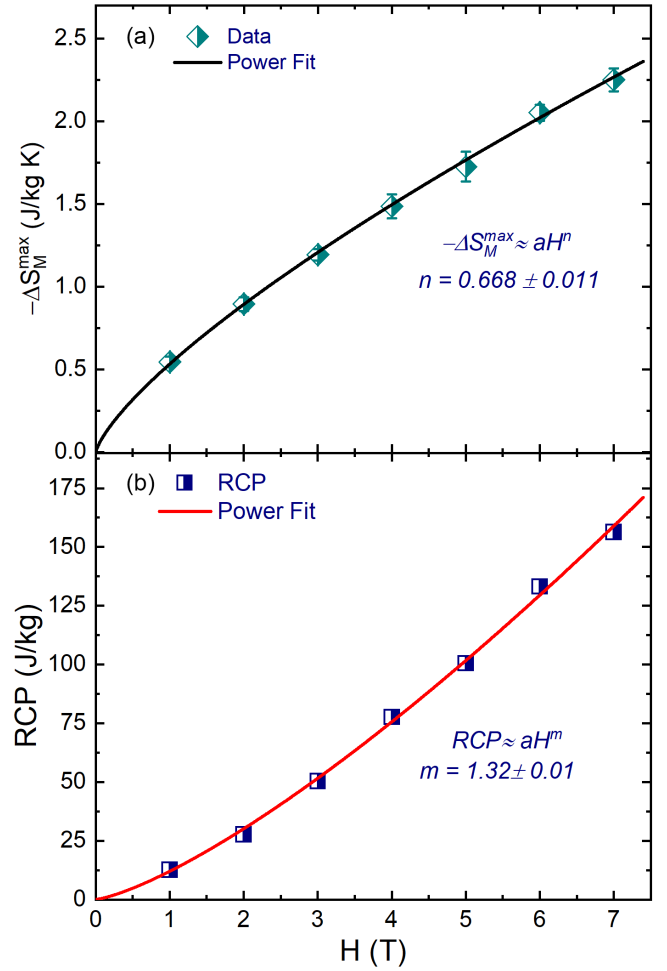


FIG. 8. (a) Field dependence of maximum magnetic entropy change ($-\Delta S_M^{\max}$) fit by a power law (solid red line). (b) Relative cooling power (RCP), with a power law fit (solid black line).

well fit by the power law $\text{RCP} \approx H^m$, with $m = 1.12(3)$. These values of RCP are comparable to or exceed those of other oxide-based magnetocaloric materials but remain below the benchmarks for room-temperature refrigeration (typically $> 300 \text{ J kg}^{-1} \text{ K}^{-1} \leq 5\text{T}$) [46,47]. However, the strong field dependence suggests that $\text{Nd}_2\text{NiMnO}_6$ and related compounds could be promising for cryogenic cooling application, where moderate RCP values ($\sim 50 - 200 \text{ J kg}^{-1} \text{ K}^{-1} \leq 5\text{T}$) are sufficient [38,41,48]. Further optimization, such as chemical substitution and/or nanostructuring may enhance their performance toward practical refrigerants.

IV. CONCLUSION

In summary, we have shown that our polycrystalline sample of the double perovskite $\text{Nd}_2\text{NiMnO}_6$ adopts a monoclinic crystal structure (space group $P2_1/n$), which hosts nearly complete B -site ordering of Ni^{2+} and Mn^{4+} cations (anti-site disorder was found to be 9%). This cation ordering was established directly by neutron powder diffraction, and confirmed through bond valence sum analysis and consideration of local distortions of the perovskite structure. Magnetometry and neutron powder diffraction measurements revealed two

magnetic phase transitions. The first at $T_1 = 198$ K corresponded to ferromagnetic ordering of the Ni^{2+} and Mn^{4+} sublattices, which was consistent with dominant ferromagnetic d - d exchange confirmed by Curie-Weiss analysis of the paramagnetic susceptibility that yielded a positive Curie-Weiss constant ($\theta_{\text{CW}} \approx 206$ K). Scaling analysis revealed that the critical phenomena at T_1 adhered to mean-field behavior characteristic of a second-order phase transition. Yet, below T_1 , ac susceptibility measurements revealed frequency-dependent behavior consistent with a reentrant spin-glass-like state. In future work, it would be interesting to quantitatively correlate the nature of low-temperature glassy behavior with the degree of antisite disorder found in this family of materials. The second transition occurred on further cooling below $T_2 = 22$ K, and was characterized by the additional noncollinear ordering of Nd^{3+} moments driven by antiferromagnetic f - d exchange and frustrated by competing antiferromagnetic f - f exchange. Symmetry analysis of this ground state magnetic structure points toward significant easy plane anisotropy of the Nd^{3+} ions. The magnetocaloric effect was characterized by determining a significant magnetic entropy change ($-\Delta S_M$) at T_1 , where the maximum change was found to be $2.25 \text{ J kg}^{-1} \text{ K}^{-1}$ under an applied field of 7 T. The respective relative cooling power was found to increase linearly with the field, stressing the potential for double perovskites such as $\text{Nd}_2\text{NiMnO}_6$ to be used in magnetic refrigeration applications. Overall, these results underpin $\text{Nd}_2\text{NiMnO}_6$ as a model system for investigating the effects of multiple competing magnetic sublattices, significant magnetocaloric effects, and possible reentrant spin-glass behavior in complex oxides.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [49].

APPENDIX A: CRITICAL POINT EXPONENTS

The critical behavior of magnetic systems near a second-order phase transition is characterized by a set of interrelated critical exponents: β , γ , and δ , which are defined as follows:

$$M(T) \propto (T_C - T)^\beta, \quad T < T_C, \quad (\text{A1})$$

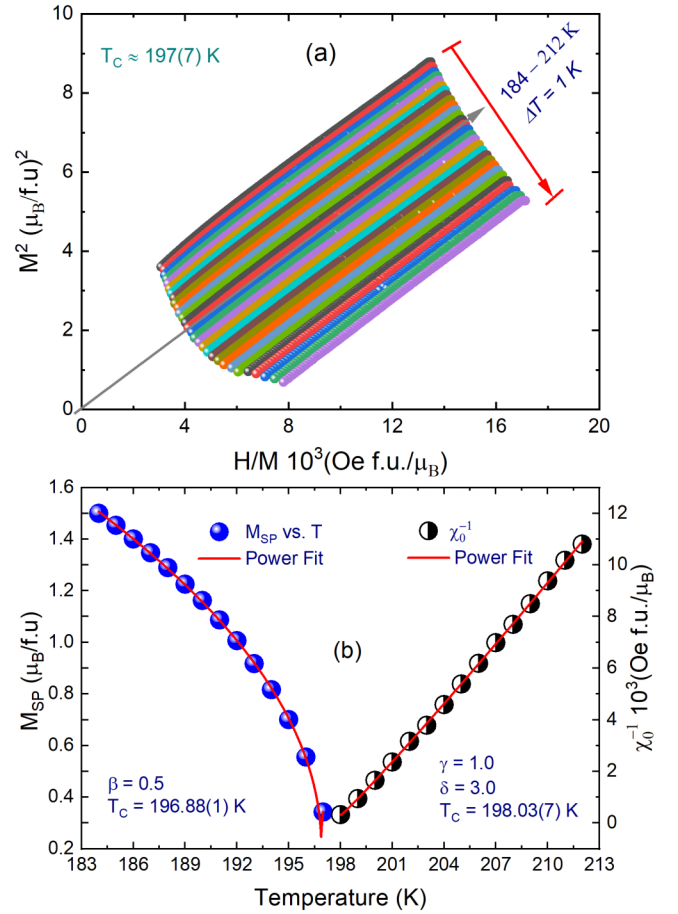


FIG. 9. (a) Arrott plot of isotherms with $\delta = 3$. (b) Temperature dependence of the spontaneous magnetization $M_S(T)$ (left) and the inverse initial magnetic susceptibility $\chi_0^{-1}(T)$ (right), obtained from the field region of $H < 1.5$ T extrapolation of the Arrott plot with $\beta = 1/2$ and $\gamma = 1$.

$$\chi_0^{-1} \propto (T - T_C)^\gamma, \quad T > T_C \quad (\text{A2})$$

$$M \propto H^{1/\delta}, \quad T = T_C. \quad (\text{A3})$$

Mean-field theories predict critical exponents of $\beta = 1/2$, $\gamma = 1$, and $\delta = 3$. In this case, Arrott plots (M^2 vs H/M), which measure the critical exponent δ , are expected to consist of a set of parallel straight line isotherms close to T_C . The Arrott plot for $\text{Nd}_2\text{NiMnO}_6$ within the temperature range of 184 – 212 K is shown in Fig. 9(a) and clearly adheres to the mean-field value of $\delta = 3$. We note that the low-field regions have been excluded, where strong curvature and deviations from linearity arise due to demagnetization effects and higher-order contributions. The value of $T_C = 197(7)$ K was determined by identifying the isotherm that intersects the origin. Figure 9(b) shows the temperature dependence of the spontaneous magnetization, $M(T)$, and the inverse magnetic susceptibility, $\chi^{-1}(T)$, obtained by extrapolating the isotherms discussed above. Fitting Eqs. (A1) and (A2) to the $T < T_C$ and $T > T_C$ regions yield T_C values of 196.88(1) K and 198.03(1) K, respectively, and provide excellent agreement with the data for mean-field values of the critical exponents.

APPENDIX B: MAGNETOCALORIC EFFECT IN $R_2\text{NiMnO}_6$ DOUBLE PEROVSKITES

In this Appendix, we summarize the magnetocaloric performance of $R_2\text{NiMnO}_6$ double perovskites (Table IV).

TABLE IV. Comparison of the magnetocaloric effect in $R_2\text{NiMnO}_6$ double perovskites. Values are given for a magnetic field change of $\mu_0\Delta H = 4.5$ T to 8 T, with those from the current work at the top.

Compound	T_C (K)	$-\Delta S_M^{\max}$ (J kg ⁻¹ K ⁻¹)	RCP (J kg ⁻¹)	Field (T)	Ref.
Nd ₂ NiMnO ₆	198	2.25	157	7	
Nd ₂ NiMnO ₆	198	1.85	125	5	
La ₂ NiMnO ₆	276	1.53		4.5	[50]
Pr ₂ NiMnO ₆	208	0.70–3.15	241.9	0–7	[51]
Pr ₂ NiMnO ₆	213–215	2.4–4.9	41.35	2–5	[40]
Nd ₂ NiMnO ₆	191	1.1–2.3	24.05	2–5	[40]
Sm ₂ NiMnO ₆	141	0.44	20.1	1.5	[52]
Eu ₂ NiMnO ₆	147	4.0	241	5	[53]
Eu ₂ NiMnO ₆	143	3.2	150	5	[54]
Gd ₂ NiMnO ₆	125	4.1	215	5	[8]
Gd ₂ NiMnO ₆	130	3.8	182	5	[54]
Tb ₂ NiMnO ₆	110	2.4–5.2	52.49	2–5	[40]
Tb ₂ NiMnO ₆	112	3.5	173	5	[54]
Dy ₂ NiMnO ₆	97	5.2	385	5	[53]
Ho ₂ NiMnO ₆	86	3.1–6.4	59.18	2–5	[40]
Y ₂ NiMnO ₆	81	2.9–5.4	74.88	2–5	[40]
Y ₂ NiMnO ₆	85	0.95–6.57		8	[38]

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