# Microscopic insights of magnetism in Sm<sub>2</sub>NiMnO<sub>6</sub> double perovskite

Supriyo Majumder <sup>(1)</sup>, <sup>1</sup> Malvika Tripathi <sup>(2)</sup>, <sup>1,2</sup> H. E. Fischer, <sup>3</sup> D. O. de Souza <sup>(2)</sup>, <sup>4</sup> L. Olivi <sup>(3)</sup>, <sup>4</sup> A. K. Sinha, <sup>5,6</sup>

R. J. Choudhary,  $^{1,*}$  and D. M. Phase  $^{1}$ 

<sup>1</sup>UGC DAE Consortium for Scientific Research, Indore 452001, India

<sup>2</sup>Niels Bohr Institute, University of Copenhagen, 2100 Copenhagen, Denmark

<sup>3</sup>Institut Laue-Langevin, 71 avenue des Martyrs, CS 20156, 38042 Grenoble cedex 9, France

<sup>4</sup>Elettra Sicrotrone Trieste S.C.p.A., SS 14-km 163.5, 34149 Basovizza, Italy

<sup>5</sup>HXAL, SUS, Raja Ramanna Centre for Advanced Technology, Indore 452013, India

<sup>6</sup>Department of Physics, School of Engineering UPES, Dehradun 248007, India

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The functional characteristics of double perovskites with a unique ferromagnetic-insulator ground state have been controversial due to the unavoidable presence of antisite disorders (ASDs). Here, we aim to investigate the origin of magnetic ordering on local and global scales in the Sm<sub>2</sub>NiMnO<sub>6</sub> (SNMO) double perovskite system. Different calcination routes are exploited to generate different cation arrangements in SNMO and the corresponding magnetic configurations are examined using the high-energy ( $E \sim 0.3 \text{ eV}$ ) "hot neutrons", which has helped to overcome Sm absorption as well as to record total (Bragg + diffuse) scattering profiles with high momentum transfer ( $Q_{\text{max}} \sim 24 \text{ Å}^{-1}$ ). We have observed that the Ni-Mn sublattice adopts long-range collinear ferromagnetic  $F_x F_z$  structure with a commensurate  $\mathbf{k} = (0, 0, 0)$  propagation vector, below ordering temperature  $T \lesssim 160$  K, irrespective of variable ASD concentrations. In addition, the signatures indicating the antiparallel polarization of Sm paramagnetic moments with respect to the Ni-Mn network are noticed in the vicinity of anomalous magnetic transitions at  $T \lesssim 35$  K. The real-space pair distribution function calculations have provided a direct visualization of ASDs by means of broadening in Ni/Mn-Mn/Ni linkage. Employing the reverse Monte Carlo approach on diffuse magnetic scattering profiles, we have observed the negative spin-spin correlation function which suggests the Ni-Ni antiferromagnetic exchange interactions ranging up to first-nearest-neighbor distance. These results confirm that the existence of ASDs in a cation ordered host matrix leads to competing ferromagnetic-antiferromagnetic phases in a broad temperature range, which quantitatively governs the temperature-dependent bulk magnetic observables of the SNMO system.

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#### I. INTRODUCTION

The existence of disorders is an unavoidable reality in every crystal system. Structural disorders are usually suspected to influence the heat conductivity [1], charge transport [2], magnetic exchange interactions [3], and many other functional properties in materials. The double perovskite oxide family  $A_2B'B''O_6$  (where, A: mainly alkaline earth or rare earth metal; B', B'': transition metal) [4], is one of the most dramatic examples of how significantly the structural dislocations can affect the electronic and magnetic ground states of the system. In double perovskites, the presence of antisite disorders (ASDs), defined as the interchange of different B', B'' cations from their respective alternate site occupancies, can even stimulate a transformation from ferromagnetic (FM)–metal (M) to antiferromagnetic (AFM)–insulator (I) as a special case [5,6].

The  $A_2$ NiMnO<sub>6</sub> (ANMO) double perovskites are the promising candidates for new-generation dissipationless quantum electronics owing to the unique FM-I ground state [7,8]. Furthermore, the near-room-temperature magnetodi-

electric, spin-pumping, magnetoresistance, and magnetocapacitance behaviors [7–9] and multiferroic properties [10,11] have aroused renewed fundamental scientific interest in the double perovskites with Ni and Mn as B-site cations. However, the nearly similar ionic radii of Ni and Mn intrinsically leads to ASDs in ANMO double perovskite systems. Noteworthily, the characteristic electronic and magnetic behaviors of ANMO systems vary significantly among different reports available in the literature [12,13]. These discrepancies can be correlated with the fact that different growth kinetics can lead to distinct cationic arrangements, which could eventually modify the physical observables [14]. As a consequence, the reliability and reproducibility related to any functional aspects of the ANMO series are questionable which limits their feasibility in device applications. Moreover, there are several unsettled fundamental issues regarding the observed physiochemical properties of the ANMO system. For instance, contradicting explanations are found about the charge state of transition metal ions participating in magnetic interactions. Goodenough et al. [15] explained the observed magnetic behavior considering Ni<sup>3+</sup>-O-Mn<sup>3+</sup> superexchange interactions, whereas Blasse et al. [16] claimed that magnetic ordering is due to Ni<sup>2+</sup>-O-Mn<sup>4+</sup> superexchange interactions. On the other

<sup>\*</sup>ram@csr.res.in

hand, both theoretical and experimental evidence suggests that the magnetic structure in the ANMO series changes considerably for different A-site ions [17,18] and can vary among collinear FM (A = La, Nd, and Y) [8,19,20], canted ferrimagnetic (A = Tb, Ho, Er, and Tm) [21], and incommensurate AFM structure (A = In) [22]. As a special case, A = Sm leads to a complicated temperature-driven magnetic phase diagram due to the unique intermixing of  $\text{Sm}^{3+}$  higher multiplet levels (J = 7/2, 9/2, etc.) with the ground state multiplet (J = 5/2)[23,24]. In the  $Sm_2NiMnO_6$  (SNMO) system, there are several discrepancies among different reports regarding the origin of the anomalous downturn in magnetic susceptibility below T = 35 K. The possible explanations of the aforementioned magnetic behavior in SNMO include (i) magnetocrystalline anisotropy governed by spin-orbit coupled Sm and Ni-Mn networks [25], (ii) long range ordering of Sm sublattice moments [12], and (iii) the emergence of a reentrant spin glass phase [26]. The presence of ASDs introduces additional perturbation in the magnetic exchange interaction pathways which causes further complexity in the ground state. Despite several research endeavors, how local ASDs can reshape the physical properties in the SNMO double perovskite system is still an open question.

One of the main reasons behind these inconsistencies is that the microscopic magnetic structure of the SNMO system and its thermal evolution are still not available in the literature. In this context, neutron diffraction is a powerful probe to study the elemental spin arrangements. On the other hand, diffuse neutron scattering could be very useful to get insight into local spin-spin correlations arising due to ASDs. However, the very high neutron absorbing  $^{149}$ Sm isotope (absorption  $\sim 42\,080$  b [27,28]) present in natural Sm creates a hindrance to perform neutron scattering experiments. The huge neutron absorption of the <sup>149</sup>Sm isotope arises possibly due to low-energy  $(\sim 0.098 \text{ eV})$  nuclear resonances [27,29]. The magnitude of neutron absorption by <sup>149</sup>Sm can be decreased significantly, if the energy of the incident neutron beam can be tuned to a value sufficiently higher than the nuclear resonance energy. Therefore, to overcome the high neutron absorption, we have utilized high-energy ( $\lambda \sim 0.5$  Å) "hot neutrons" to record diffraction profiles at the D4 disordered materials diffractometer instrument [30] at the Institut Laue-Langevin (ILL). The high momentum transfer ( $Q_{\text{max}} \sim 24$  Å<sup>-1</sup>), very low background contribution, high counting rate, and temperature stability of the D4 instrument have allowed us to record total (Bragg + diffuse) scattering patterns with good statistics. In this work, we have constructed the magnetic phase diagram of SNMO double perovskites having different degrees of cation arrangements, and explored the role of ASDs in governing the local and global magnetic structures.

## **II. EXPERIMENTAL DETAILS**

Polycrystalline SNMO bulk samples were synthesized by the solid state reaction route. Different reaction conditions are utilized to achieve distinct ASD density. Stoichiometric amounts of precursors  $Sm_2O_3$  (99.9%), NiO (99.99%), and  $MnO_2$  (99.9%) were combined to form a homogeneous mixture which was further divided into two batches. The first batch was calcined at 1200 °C for 24 h and slowly cooled at a rate  $\sim 1.6 \,^{\circ}C/min$  (sample S\_O). The next batch was calcined at 1000 °C for 24 h and thermally quenched at a rate of  $\sim 11.6 \,^{\circ}C/min$  down to  $650 \,^{\circ}C$  and then cooled at a rate of  $\sim$ 3.3 °C/min (sample S\_D). In order to identify the room temperature phase composition and the crystal structure of SNMO, powder x-ray diffraction (PXRD) and neutron powder diffraction (NPD)  $2\theta$  scans were recorded. PXRD studies were carried out using the synchrotron radiation source at the angle-dispersive x-ray diffraction (ADXRD) beamline, BL-12, Indus-II, RRCAT, Indore, India. In the PXRD measurements, the calibrated wavelength using the diffraction pattern of a LaB<sub>6</sub> standard sample was  $\lambda = 0.71949$  Å. NPD data were collected [31] utilizing the two-axis diffractometer D4 (Disordered Materials Diffractometer) [30] at ILL, Grenoble, France. The incident wavelength of the neutron beam was calibrated by recording data for the standard Ni sample and the estimated value was  $\lambda = 0.49585$  Å. D4 provides neutron diffractogram measurements covering a momentum-transfer Q range of  $\sim 0.3-23.7 \text{ Å}^{-1}$  (2 $\theta$  range  $\sim 1.3^{\circ}-138.1^{\circ}$ ) with relative Q resolution  $\sim 2.25\%$ . Temperature-dependent neutron diffraction studies were performed using a cylindrical vanadium sample container and cryostat setup. Diffraction patterns for the empty cylindrical vanadium container and cryostat assembly were collected at various temperature values. These allow us to accurately determine the temperature-dependent (if any) sample environment contributions, which is necessary for pair distribution function (PDF) analysis. Using the CORRECT program [32], the measured neutron diffraction intensity was corrected for background subtraction, sample attenuation, multiple scattering, and inelastic (Placzek) scattering effects, and normalized to absolute scale. The obtained total scattering intensity was used for PDF calculation. The crystal and magnetic structure refinement of PXRD and NPD data were performed implementing the FULLPROF package [33] wherein WINPLOTR was used for background simulation, BASIREPS was used for calculating the irreducible representations (IRs), and FPSTUDIO was used to visualize the obtained structure. A value of +5.0 fm was used for the coherent neutron scattering length of Sm at  $\lambda \sim 0.5$  Å as tabulated by Lynn and Seeger [27], from whom we also obtained the Sm absorption cross section for our neutron wavelength. The SPINCORREL and SPINVERT programs [34] based on the reverse Monte Carlo (RMC) approach were used for analyzing magnetic diffuse scattering data. The chemical valency of constituting elements was probed by x-ray absorption near-edge spectroscopy (XANES) measurements performed at room temperature in transmission mode using the synchrotron radiation source at the XAFS beamline 11.1R, Elettra-Sincrotrone, Trieste, Italy. Incident energy was calibrated by measuring absorption edges for reference Mn and Ni metal foils. The energy resolution  $\Delta E/E$  across the energy range used for XANES measurements was  $\sim 1 \times 10^{-4}$ . Standard normalization process was performed on XANES data using the ATHENA program [35]. The dc magnetization behavior was studied using the MPMS 7-tesla SQUID-VSM (Quantum Design, Inc., USA) instrument having magnetic moment sensitivity  $\sim 10^{-8}$  emu. To eliminate the trapped magnetic field of the superconducting coil inside the magnetometer, the standard *de-Gaussing* protocol was followed. To erase the prior magnetic history (if any), the sample was



FIG. 1. Rietveld analysis of (a), (b) PXRD and (c), (d) NPD  $2\theta$  scans measured at T = 300 K, showing observed (red open circles), calculated (black solid line), and difference (blue solid line) pattern along with Bragg positions (green vertical bars), for cation ordered S\_O and disordered S\_D SNMO samples. Insets of (c) and (d) show enlarged view of (011) superstructure reflection. The Rietveld generated patterns are simulated with monoclinic  $P2_1/n$  (SG 14) structural model. The goodness-of-fit indicators, for S\_O PXRD data, are  $R_p = 3.12$ ,  $R_{wp} = 4.65$ ,  $R_{exp} = 4.49$ ,  $\chi^2 = 1.07$ ; for S\_O NPD data, are  $R_p = 2.54$ ,  $R_{wp} = 2.92$ ,  $R_{exp} = 1.49$ ,  $\chi^2 = 3.82$ ; for S\_D PXRD data, are  $R_p = 7.18$ ,  $R_{wp} = 9.71$ ,  $R_{exp} = 6.43$ ,  $\chi^2 = 2.28$ ; and for S\_D NPD data, are  $R_p = 2.86$ ,  $R_{wp} = 3.75$ ,  $R_{exp} = 1.90$ ,  $\chi^2 = 3.87$ .

heated well above its magnetic transition temperatures every time before recording the data.

### **III. RESULTS AND DISCUSSION**

Room temperature powder x-ray diffractograms of SNMO samples are presented in Figs. 1(a) and 1(b). These patterns can be matched with simulated Rietveld profiles generated from both monoclinic  $P2_1/n$  (SG 14) and orthorhombic Pbnm (SG 62) structures. This is due to fact that the unit cell  $\sqrt{2a_p} \times$  $\sqrt{2a_p} \times 2a_p$ , where  $a_p$  stands for pseudocubic, is subjected to both  $P2_1/n$  and *Pbnm* space groups depending on *B*-site ordered and disordered phases, respectively. In  $P2_1/n$  symmetry there are two independent Wyckoff positions for B' and B''ions, 2c and 2d, respectively, whereas in the Pbnm symmetry, both B' and B'' ions randomly occupy identical 4b sites. Although  $P2_1/n$  is a maximal nonisomorphic subgroup of *Pbnm*, the presence of (0kl): k = 2n + 1 superstructure reflections, which is an indication for B', B'' cation ordering at alternate octahedral center sites, can be used to distinguish space group  $P2_1/n$  from Pbnm [4]. In the present case it is not easy to distinguish between  $P2_1/n$  and *Pbnm* symmetries by PXRD because (i) the mean scattering factors for x rays from Ni and Mn are nearly equal, and (ii) there is a very small difference in  $\beta$  (the angle between a and c crystallographic axes) for  $P2_1/n$  and *Pbnm*. Because of the aforementioned reasons, the superstructure peak (011) at  $2\theta \sim 9.2^{\circ}$  in PXRD having intensity 0.6% of the most intense peak at  $2\theta \sim 15.3^{\circ}$  is masked in the background contribution. On the other hand, neutron diffraction is able to probe the level of B-site cation ordering in the lattice structure owing to the distinct scattering lengths for Ni and Mn species. The room temperature NPD patterns for SNMO samples illustrated in Figs. 1(c) and 1(d) show the existence of (011) superstructure reflection at  $2\theta \sim 6.36^{\circ}$ . This confirms the dominating cation ordering present in SNMO samples and hence justifies the choice of  $P2_1/n$  space group to refine all the PXRD and NPD patterns. Noticeably, the intensity of the nuclear superstructure peak (011) with respect to the corresponding most intense peak at  $2\theta \sim$  $12.36^{\circ}$  in S O is larger in comparison to S D [Figs. 1(c) and 1(d)], which indicates qualitatively better cation ordering in S O than S D. The refined values of lattice parameters, site occupancy factors, atomic coordinates, isotropic thermal coefficients, and the reliability indicators are listed in Table I. The obtained statistical factors suggest a reasonable match between experimentally observed and simulated patterns. Good agreement between the refined parameters from PXRD and NPD scans indicates the reliability of both diffraction measurements. NPD refinement of Ni and Mn occupancies at 2c and 2d sites reveals the presence of  $\sim 3.8(6)\%$  and 11.8(8)%ASD in the S O and S D samples, respectively. As the vacancy defects may influence the electronic and magnetic properties [36], it is also important to check the possible presence of cation or anion vacancy defects in the SNMO system. The scattered x-ray intensity in PXRD measurements depends on electron density of the scattering atom; hence the contributions from heavy-Z elements dominate over low-Z elements, and any attempt to determine atomic concentration of low-Z elements such as oxygen could be erroneous. In this context, NPD is particularly advantageous as the neutron scattering cross section for the oxygen atom is large and significantly different from other constituting elements of the SNMO lattice. Therefore, we have utilized Rietveld analysis of NPD profiles measured at T = 300 K to estimate oxygen concentration in the studied SNMO samples. The obtained occupancy values as presented in Table I suggest that oxygen TABLE I. Structure parameters and reliability indicators obtained from Rietveld refinement of PXRD and NPD data recorded at T = 300 K for SNMO, S\_O: ordered and S\_D: disordered samples. Number(s) in the parentheses represent the error bars on the last digit(s). Here, the site occupancies and fractional coordinates which do not have error bars are fixed to crystallographic values.

		\$_0	S_D				
	PXRD	NPD	PXRD	NPD			
Lattice parameters (Å)							
a	5.35647(11)	5.35939(53)	5.35563(27)	5.35648(61)			
b	5.52127(11)	5.52169(49)	5.51294(26)	5.51365(52)			
с	7.61932(17)	7.61924(59)	7.61326(39)	7.61893(97)			
β	90.030(26)	90.031(12)	90.030(26)	90.030(15)			
Site occupancy &							
fractional coordinates							
Sm(4e)	1.0	1.001(5)	1.0	0.999(6)			
x	-0.00978(14)	-0.01088(80)	-0.01115(26)	-0.01107(122)			
у	0.05580(7)	0.05604(43)	0.05604(14)	0.05614(79)			
Z	0.25053(31)	0.25005(52)	0.24933(57)	0.25150(77)			
Ni Mn(2c)	0.5	0.481(1) 0.018(5)	0.5	0.440(2) 0.060(6)			
x	0.5	0.5	0.5	0.5			
у	0.0	0.0	0.0	0.0			
Z.	0.5	0.5	0.5	0.5			
Mn Ni(2d)	0.5	0.480(3) 0.020(1)	0.5	0.442(4) 0.059(1)			
x	0.5	0.5	0.5	0.5			
у	0.0	0.0	0.0	0.0			
Z.	0.0	0.0	0.0	0.0			
O1(4e)	1.0	0.994(8)	1.0	1.000(9)			
x	0.09535(103)	0.09446(51)	0.07667(203)	0.07329(68)			
y	0.47529(88)	0.47516(51)	0.47414(224)	0.47740(64)			
Z.	0.22849(139)	0.22949(36)	0.23245(196)	0.23383(44)			
O2(4e)	1.0	0.998(7)	1.0	0.991(7)			
x	0.72503(200)	0.72815(57)	0.66045(525)	0.67398(71)			
у	0.26758(171)	0.26824(40)	0.24234(328)	0.25174(73)			
Z.	0.03219(211)	0.03612(43)	0.00535(178)	0.01260(54)			
O3(4e)	1.0	0.994(9)	1.0	1.004(8)			
x	0.69691(175)	0.70041(53)	0.69099(172)	0.69126(61)			
у	0.29364(124)	0.29330(55)	0.32438(199)	0.32333(63)			
Z.	0.45827(147)	0.45825(56)	0.43036(159)	0.43290(44)			
Isotropic thermal factors (Å <sup>2</sup> )							
B <sub>Sm</sub>	0.072(9)	0.100(31)	0.068(17)	0.121(45)			
B <sub>Ni</sub>	0.049(39)	0.089(17)	0.057(28)	0.091(27)			
B <sub>Mn</sub>	0.046(39)	0.123(51)	0.054(24)	0.110(79)			
Statistical parameters							
$R_p$	3.12	2.54	7.18	2.86			
$\dot{R}_{wp}$	4.65	2.92	9.71	3.75			
R <sub>exp</sub>	4.49	1.49	6.43	1.90			
$\chi^2$	1.07	3.82	2.28	3.87			

stoichiometry is maintained in SNMO systems. Empirically calculated values of the Goldschmidt tolerance factor [37] are  $t \approx 0.929$  for Ni<sup>2+</sup>, Mn<sup>4+</sup> valence states; 0.932 for Ni<sup>3+</sup> lowspin (LS), Mn<sup>3+</sup> high-spin (HS) states; and 0.923 for Ni<sup>3+</sup> HS, Mn<sup>3+</sup> HS states. The t < 1 situation leads to tilting of (Ni/Mn)O<sub>6</sub> octahedra which causes distortion in the system.

Absorption spectra across Mn and Ni *K* edges measured for SNMO samples along with standard references for  $Mn^{4+}$ (MnO<sub>2</sub>),  $Mn^{3+}$  (Mn<sub>2</sub>O<sub>3</sub>), Ni<sup>2+</sup> (NiO), and Ni<sup>3+</sup>(Ni<sub>2</sub>O<sub>3</sub>) are depicted in Figs. 2(a) and 2(b). SNMO XANES spectra for both Mn and Ni *K* edges show a characteristic white line just near the absorption edge along with pre-edge features and post-edge oscillatory behavior. XANES spectra at the transition metal (TM) K edge is related to photoelectron excitation from atomic 1s to empty p bands. The pre-edge features in XANES spectra may arise because of 1s-3d quadrupole or/and weakly allowed dipole transitions. TMO<sub>6</sub> octahedral distortions give rise mixed p-d states which cause such weakly allowed s-d transition [38]. The multiple scattering contributions from different coordination shells result in postedge oscillations. It is known that as the valency of the TM ion increases, the band edge which is defined as the first inflection point in absorption spectra, shifts toward higher energy values [39]. Comparing the observed K edge XANES spectra [insets of Figs. 2(a) and 2(b)], it can be inferred that (i) both of the SNMO samples have almost the same absorption



FIG. 2. (a) Mn and (b) Ni K edge XANES spectra measured at T = 300 K for SNMO samples along with standard references. Insets show first derivative of normalized absorption with respect to incident photon energies.

edge energy for Mn and Ni species, and (ii) Mn and Ni ions present in SNMO samples have valency in between 3+, 4+ and 2+, 3+, respectively. The observed mixed valence nature of both TM species indicates charge disproportion between *B*site cations through  $Ni^{2+} + Mn^{4+} \rightarrow Ni^{3+} + Mn^{3+}$ , which is also reported in many other double perovskite systems [8,40]. With varying ASD we have not observed apparent changes in spectral characters in XANES, suggesting charge states of constituting elements remain unbiased to ASD densities.

The dc magnetometric response acquired for SNMO samples is shown in Fig. 3. The temperature dependency of the magnetic moment M(T) measured in typical zero-fieldcooled (ZFC) warming and field-cooled warming (FCW) protocols under 25 Oe of applied magnetic field reveals two distinct magnetic transitions, (i) at  $T = T_C$ , defined as the onset temperature point of M(T), and (ii) at  $T = T_d$ , defined as the downturn in M(T). The transition temperature  $T_C$ is determined from the first temperature derivative of FCW magnetization dM/dT [inset (i) of Fig. 3(a)] and found to be  $T_C = 159.6 \text{ K}$  for S\_O and  $T_C = 156.6 \text{ K}$  for S\_D.  $T_d$  is estimated from the temperature where M(T) attains the maximum value and is approximated to be  $T_d \sim 34.1 \,\mathrm{K}$  for both S\_O, S\_D samples. The overall magnetic moment decreases with increasing ASD concentrations. In the dM/dT curve, a broad inverted cusplike behavior at around  $T \sim 67 \,\mathrm{K}$  is observed which is more prominent in the S D sample than in S\_O, as illustrated in Fig. S1 in the Supplemental Material (SM) [41]. This feature can be attributed to Mn-O-Mn shortscale interaction due to antisite disordered bonds [42]. M(T)recorded under different measuring magnetic field values, as shown in Fig. 3(b) for the S\_O sample, indicates robustness of transition temperature  $T_C$ . Below  $T_C$  a clear thermal hysteresis in between field-cooled cooling (FCC) and warming magnetization paths having broad temperature width of  $\Delta T \sim 140 \,\mathrm{K}$ in the presence of  $\mu_0 H = 25$  Oe is observed for the S\_O sample as displayed in Fig. 3(b). With increasing applied field strength ( $\mu_0 H\gtrsim 1$  kOe) the thermal hysteresis vanishes. Similar behavior is also confirmed for the S\_D sample (data not shown here). The downturn transition shifts toward the lowtemperature side with increasing measuring field values (at  $\mu_0 H \sim 30$  kOe) and with a further increase of magnetic field  $(\mu_0 H > 30 \text{ kOe})$  the downturn behavior seems to transform into an upturn as shown in Fig. 3(c), examined for the S\_O sample. A qualitatively similar trend is observed for the S\_D case (data not shown here) also. The isothermal magnetization M(H) measured at T = 5 K, presented in Fig. 3(d), reveals a decrease in saturation moment value with increasing ASD in the SNMO system. Experimentally estimated moment  $\mu_{\text{expt}}$ values at  $\mu_0 H = 70 \text{ kOe}$ , T = 5 K are found to be 5.56  $\mu_B$  in S\_O and 5.21  $\mu_B$  in S\_D. M(H) at T = 300 K shows typical paramagnetic (PM) behavior.

Figures 4(a) and 4(b) depict the contour maps of temperature-dependent neutron scattering intensity in the low-Q regime measured for SNMO samples. Transition temperatures obtained from the aforementioned bulk magnetometric results are marked by vertical dashed  $(T_C)$  and dotted  $(T_d)$  lines. With change in temperature, distinguishable variations in measured neutron counts are observed at  $Q \sim 1.6$ and 2.3 Å<sup>-1</sup> in the vicinity of  $T_C$  and  $T_d$  transitions. In order to have better visualization, measured temperature-dependent NPD profiles are vertically stacked as shown by Figs. S2(a) and S2(b) in the SM [41]. The comparison of NPD patterns recorded at  $T = 12 \text{ K} (T < T_C)$  and  $T = 180 \text{ K} (T > T_C)$  for SNMO samples is highlighted in Figs. 5(a) and 5(b). Pointby-point difference spectra between these two temperatures are evaluated to eliminate the scattering contributions from nuclear structure and background. Temperature-dependent PXRD analysis (not shown here) confirms the absence of a symmetry-related structural phase transition within the investigated temperature region (5 K  $\leq T \leq$  300 K). Therefore, the difference patterns present magnetic scattering along with a temperature-driven effect (Debye) on nuclear reflections. The negative values in difference spectra arise from dominating diffuse scattering contributions at T = 180 K superimposed underneath the Bragg reflections. For both SNMO samples, no new magnetic peak emerges. Magnetic reflections are contributed by means of intensity enhancement at same nuclear Bragg peak positions. Hence, in SNMO the magnetic and nuclear unit cells coincide and  $\mathbf{k} = (0, 0, 0)$  is chosen as the propagation vector for the magnetic structure. Propagation vector  $\mathbf{k} = (0, 0, 0)$  is also used to define the magnetic unit cell in other members of the ANMO family [19-21]. The



FIG. 3. (a) Magnetization as a function of temperature M(T) measured under  $\mu_0 H = 25$  Oe for SNMO ordered S\_O and disordered S\_D samples. Inset shows temperature derivative of field cooled warming cycles of M(T). Effect of different measuring magnetic field values on (b) (left panel) thermal hysteresis in cooling-warming magnetization cycles, (b) (right panel) PM to FM transition temperature  $T_C$ , and (c) low-temperature downturn transition  $T_d$ , examined for S\_O sample. (d) Isothermal magnetic hysteresis loops for SNMO samples at T = 5 K and 300 K.

appearance of magnetic peaks at the same nuclear Bragg positions indicates the FM nature of SNMO samples. At lower  $2\theta$  values of the NPD profile distinguishable changes are prominently observed in peak intensity against temperature variation. Such abrupt change in peak intensity with temperature suggests that these low-angle reflections have dominating magnetic characters over nuclear contribution. The peak at  $2\theta \sim 7.44$  Å marked as "A" is indexed by closely spaced

Bragg reflections  $(110)_{n+m}$  and  $(002)_{n+m}$ , where *n* and *m* correspond to nuclear and magnetic contributions, respectively. The reflection at  $2\theta \sim 10.57$  Å marked as "B" is indexed by closely spaced  $(020)_{n+m}$ ,  $(112)_{n+m}$ , and  $(200)_{n+m}$  Bragg peaks. The thermal evolution of features A and B as shown in Figs. 5(c) and 5(d) behaves like the magnetic order parameter. On lowering temperature both A and B features reveal onset in intensity at around  $T = T_C$ . On further decrease of tempera-



FIG. 4. Contour map of temperature-dependent measured neutron intensity in low-Q region for SNMO (a) ordered S\_O and (b) disordered S\_D samples. The vertical dashed and dotted lines highlight  $T_C$  and  $T_d$ , respectively, derived from magnetometric measurements.



FIG. 5. (a), (b) NPD patterns collected at T = 12 K, 180 K along with corresponding temperature subtracted difference spectra for SNMO ordered S\_O and disordered S\_D samples. Insets of (a) and (b) show enlarged view of low-angle peaks A:  $(110)_{n+m} + (002)_{n+m}$  and B:  $(020)_{n+m} + (112)_{n+m} + (200)_{n+m}$ , where *n*, *m* correspond to nuclear and magnetic contributions, respectively. Temperature dependency of NPD intensity at feature A and B 2 $\theta$  positions for (c) S\_O and (d) S\_D samples. Open symbols are for observed data and solid lines are guides to the eyes. (e) Best-fit curves along with experimentally observed NPD diffractograms at T = 5 K across A and B reflections modeled with different IRs for S\_O sample.

ture at  $T \sim T_d$  intensity of feature B encounters a downturn whereas intensity of A increases. In a previous study [43], the cation disordered (*Pbnm* symmetry) Ho<sub>2</sub>FeCoO<sub>6</sub> double perovskite is found to show significant intensity redistribution in magnetic Bragg lines at low temperature, which is identified as a spin reorientation transition leading to transformation from mixed magnetic structure to single magnetic structure. However, this is not the case for the SNMO system, as here a single magnetic structure is evidenced for all temperature values below  $T = T_C$  (discussed later). Therefore, the observed magnetic intensity redistribution indicates temperature-driven changes in the magnetic structure of SNMO samples.

The  $P2_1/n$  space group symmetry allowed possible magnetic structures for the  $\mathbf{k} = (0, 0, 0)$  propagation vector are computed with the help of representation theory analysis developed by Bertaut [44,45]. The obtained IRs and associated

basis vectors for the Sm, Ni, and Mn sublattices are listed in Table II. Different trial magnetic representations are tested in the refinement model and the corresponding quality of fitting is checked. As presented in Fig. 5(e), the best fit to experimental pattern is obtained for IR(3).

Rietveld refinements of NPD data at T = 12 K, presented in Figs. 6(a) and 6(b), show good agreement between experimental and calculated patterns. The obtained spin arrangements at T = 12 K, as illustrated in Figs. 6(c) and 6(d), reveal the  $F_xF_z$  magnetic structure of the Ni-Mn network. Moment vectors are found to lie in the *a*-*c* plane. The nonzero moment component along the *b* axis does not improve the fitting quality and even diverges. Therefore, magnetic structure analysis unambiguously confirms that below  $T = T_C$  SNMO undergoes collinear FM ordering of Ni, Mn magnetic sublattices. The thermal variation of element-specific magnetic

TABLE II. SNMO magnetic structure character table generated for  $P2_1/n$  space group with  $\mathbf{k} = (0, 0, 0)$  propagation vector.

	IR matrices in symbolic form Symmetry operators								Basis vectors Atomic sites								
IRs	1	2 (0,	1/2, 0)	1/4,	, y, 1/4 -	-1 0,0,0	<i>n</i> (1/2, 0,	1/2) x, $1/4$	4, <i>z</i>	Sm1	Sm2	Sm3	Sm4	Ni1	Ni2	Mn1	Mn2
IR(1)	1		1			1		1	(1	(u, v, w)	(-u, v, -w)	(u, v, w)	(-u, v, -w)	(r, s, t)	(-r, s, -t)	(o, p, q)	(-o, p, -q)
IR(2)	1		1			-1		-1	(1	(u, v, w)	(-u, v, -w)	(-u, -v, -w)	(u, -v, w)				
IR(3)	1		-	1		1		-1	(1	(u, v, w)	(u, -v, w)	(u, v, w)	(u, -v, w)	(r, s, t)	(r, -s, t)	(o,p,q)	(o, -p, q)
IR(4)	1		-	1		-1		1	(1	(u, v, w)	(u,-v,w)	(-u,-v,-w)	(-u, v, -w)				



FIG. 6. (a), (b) Rietveld analysis of NPD  $2\theta$  scans recorded at T = 12 K showing observed (red open circles), calculated (black solid line), and difference (blue solid line) pattern along with Bragg positions (green vertical bars) for SNMO ordered S\_O and disordered S\_D samples. The Rietveld generated patterns are simulated with monoclinic  $P2_1/n$  (SG 14) nuclear structure and IR(3) magnetic structure with  $\mathbf{k} = (0, 0, 0)$  propagation vector. The goodness-of-fit indicators, for S\_O data, are  $R_p = 1.73$ ,  $R_{wp} = 1.92$ ,  $R_{exp} = 0.99$ ,  $\chi^2 = 3.78$ ; and for S\_D data, are  $R_p = 2.21$ ,  $R_{wp} = 2.60$ ,  $R_{exp} = 1.07$ ,  $\chi^2 = 5.87$ . Refined magnetic structures at T = 12 K for (c) S\_O and (d) S\_D samples.

moments from Sm, Ni, and Mn sublattices are presented in Figs. 7(a) and 7(b). The temperature evolution of magnetic moment components along the a(x) and c(z) directions obtained from Rietveld refinement is shown in the insets of Figs. 7(a) and 7(b). With decreasing temperature, Ni and Mn spins develop long-range ordering in the FM configuration and corresponding sublattice magnetic moment values start increasing below  $T = T_C$ . We have not observed any signature of Sm sublattice long-range ordering within 5 K  $\leqslant$  T  $\leqslant$ 300 K. However, PM Sm moments are polarized due to the internal exchange field from Ni-Mn ordered sublattice. Upon further cooling across  $T = T_d$  polarized Sm moments align opposite to Ni-Mn moments, as illustrated in Figs. 7(c)-7(h). With increasing ASD concentration the magnetic moment values are found to decrease as depicted in Figs. 7(a) and 7(b). This trend is in agreement with magnetometric observation.

Sine Fourier transformation of corrected neutron total scattering intensity S(Q) is calculated as [46,47]

$$G(r) = K(2/\pi) \int_{Q_{\min}}^{Q_{\max}} [S(Q) - 1]Q\sin(Qr)dQ,$$

where *r* is atomic pair separation distance, *K* is absolute scale normalization factor, and S(Q) has been normalized to 1 at high *Q* after subtraction of incoherent scattering from the diffraction pattern. Figures 8(a) and 8(b) present a comparison of SNMO samples having different ASDs in the low-*r* region of calculated real-space radial distribution histograms at T = 300 K and 12 K, respectively. PDF is an ideal probe for local structural modifications and cationic arrangement because it considers both Bragg and diffuse scattering on an equal footing. In the nuclear PDF profile, the peak positions contain information about all possible atom-atom coordination distances. Ideally, each atom pair should contribute to a well-defined delta function, whereas in real systems, the distribution of interatomic separations due to disorder causes peak broadening. The peak heights depend on scattering factors of atomic pairs. The integrated intensity of a given peak corresponds to coordination numbers of atom pairs [48,49]. At T = 300 K, as SNMO is PM [Fig. 3(d)], the dominating contribution in observed PDF comes from nuclear (Bragg + diffuse) scattering. Different regions of these PDF are identified from refined nuclear structure displayed in Fig. 8(c). Noticeable differences are observed in PDF histograms of SNMO samples with different ASD densities across some specific regions, where B-site (Ni/Mn) atoms are involved. Particularly, the feature around  $r \sim 3.9$  Å which corresponds to Ni/Mn-Mn/Ni atomic pair contribution becomes wider in S\_D than S\_O, as highlighted in Figs. 8(a) and 8(b). More peak broadening again confirms the presence of more ASD in S\_D as compared to S\_O. With decreasing temperature PDF features become more prominent and narrower, as shown in Figs. S4(a) and S4(b) in the SM [41]. This is because, in low-temperature PDF, (i) pair correlations are strong as magnetic (Bragg + diffuse) scattering also has significant contribution along with nuclear scattering, and (ii) with reducing temperature the amplitude of thermal vibrations in atom pairs is reduced. Comparison of PDF at T = 12 K as presented in Fig. 8(b) shows much more broadening than the



FIG. 7. Temperature dependency of element-specific sublattice moments for SNMO (a) ordered S\_O and (b) disordered S\_D samples. Insets of (a) and (b): Variation of moment components along x (*a*-axis) and z (*c*-axis) directions. Geometric symbols are for values obtained from refinements and solid lines are guides to the eyes. Spin arrangement for S\_O sample in (c), (e), (g) *a-b* plane and (d), (f), (h) *a-c* plane, obtained by refining NPD data recorded at T = 5 K, 45 K, and 75 K, respectively. The Rietveld fitting of NPD 2 $\theta$  scans recorded at T = 5 K, 45 K, and 75 K, are presented in Fig. S3 in the SM [41]. Goodness-of-fit indicators, for T = 5 K data, are  $R_p = 2.03$ ,  $R_{wp} = 2.23$ ,  $R_{exp} = 1.45$ ,  $\chi^2 = 2.37$ ; for T = 45 K data, are  $R_p = 1.59$ ,  $R_{wp} = 1.77$ ,  $R_{exp} = 0.92$ ,  $\chi^2 = 3.73$ ; and for T = 75 K data, are  $R_p = 1.72$ ,  $R_{wp} = 1.94$ ,  $R_{exp} = 0.98$ ,  $\chi^2 = 3.91$ .

T = 300 K case for Ni/Mn-Mn/Ni pair correlation features with increasing ASD. This again indicates weaker magnetic long-range (Bragg) and stronger diffuse (nuclear + magnetic) scattering due to more ASD present in the S D sample.

We have already observed the signature of diffuse magnetic scattering contributions superimposed underneath the Bragg reflections at T = 180 K [Figs. 5(a) and 5(b)]. In order to extract the diffuse magnetic scattering contribution, the NPD pattern collected at T = 300 K is subtracted from the T = 180 K pattern as presented in the inset of Fig. 8(d). This temperature subtraction eliminates the nuclear Bragg scattering contribution from T = 180 K data. The cation ordered SNMO lattice can be considered as an alternate stacking arrangement of SmNiO<sub>3</sub> and SmMnO<sub>3</sub> single perovskites, whereas in the cation disordered SNMO lattice, SmNiO<sub>3</sub> and SmMnO<sub>3</sub> have random distribution. Therefore, in the SNMO system, magnetic interactions are possible in between Sm-Sm, Sm-Ni, Sm-Mn, Ni-Ni, Ni-Mn, and Mn-Mn spin pairs via intermediate O ions. Due to long separation between cation pairs, superexchange through intermediate O ions dominates over direct exchange interaction between spins. Generally a rare earth metal orders at very low temperature (as here, no signature of Sm ordering within 5 K  $\leq T \leq 300$  K) and has relatively very small moment [as observed from Figs. 7(a) and 7(b)] values in comparison to TM ions. Hence in diffuse magnetic scattering study, interactions from spin pairs involving Sm ions can be neglected. The long-range FM ordering of Ni-Mn sublattices in SNMO are observed below

 $T_C \simeq 160$  K. In SmMnO<sub>3</sub> [42] and SmNiO<sub>3</sub> [50] perovskites, Mn-Mn and Ni-Ni moments are reported to order in AFM configurations, below  $T_N \simeq 59$  K and 225 K, respectively. Therefore, the temperature-subtracted scattering intensity at T = 180 K should have a dominating contribution from Ni-Ni spin pairs. The RMC approach on a  $10 \times 10 \times 10$  supercell filled with randomly oriented classical spin vectors (4000 spins) at B-site sublattices obeying periodic boundary conditions is employed to analyze diffuse magnetic scattering. Spin rotation at random sites is performed following algorithm acceptance criteria until the best possible fitting of observed data is achieved. The RMC fit of diffuse magnetic scattering data using Ni magnetic form factor and obtained real-space spin-spin correlation distribution at T = 180 K are presented in Fig. 8(d). Here the radial spin-spin correlation distribution function  $\langle S(0) \cdot S(r) \rangle$  represents average scalar product of spin pairs situated at center r = 0 and at a distance r. The negative value of spin correlation function at the nearest-neighbor site ( $r \sim 3.8$  Å) unambiguously confirms the presence of short-scale Ni-Ni AFM interactions in SNMO. Such short-scale coupling originates from ASD Ni-O-Ni bonds in SNMO.

In the present scenario, due to the coexisting of ordered, disordered phases and mixed valency of *B*-site cations, a number of magnetic exchange interactions are possible in the SNMO system: (i) FM superexchange interaction through Ni<sup>2+</sup>( $e_g^2$ )-O-Mn<sup>4+</sup>( $e_g^0$ ) in 180° geometry from cation ordered configurations [51,52]; (ii) AFM interaction because



FIG. 8. Real-space PDF histograms at (a) T = 300 K and (b) T = 12 K for SNMO ordered S\_O and disordered S\_D samples. Features for Ni/Mn-Mn/Ni atomic pairs are highlighted by box. (c) Refined nuclear structure for SNMO system, demonstrating different interatomic separation distances in Å: (i) Ni-O1: 1.819(4), Ni-O2: 1.960(3), Ni-O3: 1.968(4); (ii) Mn-O1: 2.132(4), Mn-O2: 1.938(3), Mn-O3: 1.995(4); (iii) Sm-O1 short: 2.393(3), long: 4.221(6), 4.493(5), 4.504(6), Sm-O2 short: 2.559(5), long: 4.415(5), 4.437(6), 4.578(5), Sm-O3 short: 2.576(6), long: 4.328(6), 4.486(5), 4.539(5); (iv) O1-O2: 2.638(5), O1-O3: 2.924(5); (v) Sm-Ni: 3.106(4), 3.257(5), 3.345(5), 3.613(4); (vi) Sm-Mn: 3.105(4), 3.257(5), 3.348(5), 3.612(4); (vii) Sm-Sm: 3.771(7); (viii–x) Ni/Mn-Mn/Ni: 3.8093(4), 3.8470(3). Here, the number in parentheses represents the error bars on the last digit. Ni-Mn pair belongs to *B*-site cation ordered structures whereas Ni-Ni and Mn-Mn pairs are associated with cation disordered structures. (d) Radial spin-spin correlation distribution obtained from fitting diffuse magnetic scattering data (inset) at T = 180 K for S\_D sample. In inset red open circles are for observed, black solid line is for calculated, and green solid line is for difference pattern.

of virtual charge transfer between Ni<sup>2+</sup>( $e_g^2$ )-O-Ni<sup>2+</sup>( $e_g^2$ ) and Mn<sup>4+</sup>( $t_{2g}^3$ )-O-Mn<sup>4+</sup>( $t_{2g}^3$ ) ASD pairs; (iii) FM interaction due to vibronic superexchange between  $Ni_{LS}^{3+}(e_g^1)$ -O-Mn $_{HS}^{3+}(e_g^1)$ (where LS: low-spin state, HS: high-spin state) cation ordered pairs; (iv) AFM superexchange interaction between ASD pairs via  $Ni_{LS}^{3+}(e_g^1)$ -O-N $i_{LS}^{3+}(e_g^1)$  and  $Mn_{HS}^{3+}(e_g^1)$ -O-M $n_{HS}^{3+}(e_g^1)$ ; (v) exchange interaction between all combinations of Ni and Mn pairs, when  $Ni^{3+}$  stabilizes in the HS state [53]. (vi) AFM superexchange interaction in 180° linkage between  $Ni^{2+}(e_g^2)$ -O-Ni<sup>3+</sup> $(e_g^1)$ . (vii) FM double exchange interaction between Mn<sup>3+</sup>( $e_{\sigma}^{1}$ )-O-Mn<sup>4+</sup>( $e_{\sigma}^{0}$ ). In addition, these aforementioned magnetic exchange interactions can be affected by local octahedral distortion, since both Ni<sup>3+</sup> and Mn<sup>3+</sup> have orbital degeneracy and are Jahn-Teller active. As observed in present case, SNMO samples have significant amount of cation ordered phase (cation ordering in S\_O is ~96.1% and in S\_D is ~86.5%) and hence magnetic interactions (i) and (iii) have dominating contribution. However, the Ni<sup>3+</sup><sub>LS</sub>-O-Mn<sup>3+</sup><sub>HS</sub> vibronic superexchange interaction (iii) is less stable in comparison with static superexchange interaction (i) through Ni<sup>2+</sup>-O-Mn<sup>4+</sup>. With increasing ASD densities, contributions from the (ii) and (iv) interactions are enhanced. The

phase boundary effects through interactions (vi) and (vii) are relatively feeble in comparison with contributions from ordered or disordered regions. As a consequence of these coexisting exchange pathways, the magnetic behavior of SNMO comprises competing FM-AFM interactions, which are respectively originated from the *B*-site ordered and disordered structures. The observed transition at  $T = T_C$  in the SNMO system is assigned to long-range FM ordering from Ni-O-Mn *B*-site ordered chains. The theoretical effective magnetic moments for different spin configurations defined as  $\mu_{\text{theo}} = \sqrt{2 \times \mu_{\text{Sm}}^2 + \mu_{\text{Ni}}^2 + \mu_{\text{Mn}}^2}$ , where  $\mu_{\text{Sm}} = g\sqrt{J(J+1)}$ with  $g \simeq 0.2857$  and  $\mu_{\text{Ni}/\text{Mn}} = g\sqrt{S(S+1)}$  with  $g \simeq 2.00$ , yield the following values:  $4.94 \,\mu_B$  for [Sm<sup>3+</sup> (J = 5/2), Ni<sup>2+</sup> (S = 1), Mn<sup>4+</sup> (S = 3/2)]; 6.36  $\mu_B$  for [Sm<sup>3+</sup> (J = 5/2), Ni<sup>3+</sup><sub>HS</sub> (S = 3/2), Mn<sup>3+</sup><sub>HS</sub> (S = 2)]; and 5.33  $\mu_B$  for [Sm<sup>3+</sup> (J = 5/2), Ni<sup>3+</sup><sub>LS</sub> (S = 1/2), Mn<sup>3+</sup><sub>HS</sub> (S = 2)].

Therefore, the observed  $\mu_{expt}$  values suggest mixed valence spin interaction of *B*-site cations. XANES measurements for SNMO samples suggest that mixed valence nature of both Ni and Mn ions remains unbiased to ASD concentration. Therefore, the smaller  $\mu_{expt}$  value of S\_D than that of S\_O is solely due to different level of cation ordering. ASD mediated shortscale AFM couplings through Ni-O-Ni or Mn-O-Mn bond pairs weaken the predominant FM interactions in the ordered host matrix. Consequently, increase of ASD concentration results in reduction of net magnetic moment and saturation magnetization. This decrease of moment values with increasing ASD concentration is confirmed from magnetometric measurements as well as NPD studies. With increasing ASD density short-range correlations increase in the system and this leads to more prominent inverted cusplike broad transition in the S\_D sample as compared to the S\_O case. The thermomagnetic irreversibility in FCC and FCW magnetization cycles in any system can be due to magnetic frustration [54], or first-order phase transition [55], or a spin-glass-like state or domain wall pinning, or arrested dynamic behavior [56]. In the present case, the observed thermal hysteresis in SNMO samples possibly originates from frustration due to competing exchange interactions in mixed magnetic phases. At high measuring field values ( $\mu_0 H \gtrsim 1$  kOe) the long-range magnetic interaction from the ordered phase dominates over the diluted short interaction in the disordered phase and consequently the thermomagnetic irreversibility vanishes. The sharp reduction in magnetization below  $T = T_d$  suggests the possibility of antiparallel alignment of constituent moments in the magnetic lattice. During cooling from the high-temperature PM state below  $T_C$ , Ni and Mn ions order in FM configuration and impose an internal magnetic field H<sub>int</sub> at Sm sites. Experiencing the internal exchange field effect exerted by the long-range ordered Ni-Mn sublattice, PM Sm moments are polarized in the antiparallel direction with respect to the Ni-Mn network. The opposite alignment of polarized rare-earth moments with respect to TM moments due to the presence of an internal magnetic field has been observed in some perovskite and double perovskite structures [19,57,58]. With decreasing temperature, there is increase of orientational ordering of polarized Sm PM moments. At  $T = T_d$  the competition between the oppositely aligned overall magnetic moment of the Ni-Mn sublattice and polarized PM moment of the Sm site results in downturn behavior in temperature-dependent magnetization. When the measuring field balances the internal field (therefore,  $H_{\rm int} \simeq 30$  kOe) there is no longer any decrease in magnetic moment with decreasing temperature.

### **IV. CONCLUSION**

In summary, we have addressed the microscopic magnetic configuration of the Sm<sub>2</sub>NiMnO<sub>6</sub> double perovskite system. Depending on the calcination route, SNMO samples crystallize with differing cation arrangements but in the same monoclinic  $P2_1/n$  structure. We have confirmed that the mixed valence nature of both Ni and Mn species arising due to Ni<sup>2+</sup> + Mn<sup>4+</sup>  $\rightarrow$  Ni<sup>3+</sup> + Mn<sup>3+</sup> charge disproportion is independent with respect to variation of antisite disorder concentration. High-energy ( $\lambda \sim 0.5$  Å) neutrons are utilized to overcome the high neutron absorption of natural Sm ions and to record the thermal variation of diffraction patterns. The degree of antisite disorders is quantified by refining the intensity corresponding to nuclear Bragg reflections. Below the magnetic ordering temperature (T < 160 K), the moments at Ni and Mn sites follow long-range collinear ferromagnetic arrangement with spin components along the a and ccrystallographic axes, irrespective of different antisite disorder extents. Thermal evolution of individual ionic magnetic moments indicates the opposite alignment of polarized Sm moments with respect to Ni-Mn sublattice moments below  $T \sim 35$  K. Pair distribution function calculations depict the decrement in magnetic Bragg intensity, but the enhancement in diffuse (nuclear + magnetic) scattering with increasing the degree of antisite disorders. Reverse Monte Carlo analysis of diffuse magnetic scattering profiles reveals the existence of antisite disorder mediated Ni-Ni short range antiferromagnetic couplings ranging up to first-nearest-neighbor distances, which sustain even above the ferromagnetic ordering temperature. As a consequence of these additional Ni-O-Ni (or Mn-O-Mn) local antiferromagnetic interactions in the predominantly ordered Ni-O-Mn ferromagnetic host matrix, the magnetic behavior of SNMO comprises coexisting magnetic phases in a wide temperature regime. The present study will in general provide a route map to control the characteristic magnetic aspects by properly tuning the antisite disorders in all the prototypical double perovskite systems.

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