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5 **Effects of neodymium doping on the structural and magnetic properties**
 6 **of the double perovskite $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ compound**

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21 Curie temperature (T_C) of the ferromagnetic compound $\text{Sr}_2\text{FeMoO}_6$ can be modified by
 22 increasing the itinerant electrons at the Fermi level. Nd doping increases the electron
 23 density but the charge neutrality should be taken into account. In this work, a series of
 24 double perovskites $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ ($y = 0.0, 0.25, 0.5, 0.75$ and 1.0) for both
 25 cases, the off-stoichiometric ($x = 0$) and the stoichiometric ($x > 0$ with $x = y/2$) one,
 26 were synthesized via solid state route and discussed in detail. A crystalline structure
 27 transition was confirmed by X-ray diffraction analysis, showing a change of the lattice
 28 parameters and the unit cell volume as a function of Nd doping. It was also found
 29 that the nominal valence in Fe is mainly Fe^{3+} and in Mo is Mo^{4+} . The T_C increases
 30 continuously with Nd doping in the stoichiometric case. The increase of the T_C follows a
 31 competition with the substitution of Sr^{2+} ions by Nd^{3+} ions and the charge neutrality
 32 considered in our system.

33 *Keywords:* **Double perovskite, Nd doping, Curie Temperature.**

34 **1. Introduction**

35 The ferromagnetic oxide $\text{Sr}_2\text{FeMoO}_6$ (SFMO) with a double perovskite structure
 36 and strong electron correlation is considered as a fundamental compound to under-
 37 stand the role of electronic and magnetic properties in half-metallic systems.¹

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1 SFMO has been investigated in the past decade due to attractive properties and the
 2 wide range of potential applications in spintronics² such as recording devices and
 3 magnetic sensors. This is attributed to the peculiar properties of SFMO, full spin
 4 polarization, low-field magnetoresistance and high Curie temperature (T_C). In the
 5 fully ordered structure of the $\text{Sr}_2\text{FeMoO}_6$ compound, Fe and Mo atoms occupy two
 6 interpenetrating sublattices α and β , respectively. Oxygen ions bridge the Fe and
 7 Mo atoms to form alternating FeO_6 and MoO_6 octahedra. Experimental results of
 8 the spin polarization give values of $P \approx 0.85$,³ close to the theoretical full polariza-
 9 tion value $P = 1$ reached for the saturation magnetization with an expected value
 10 of $4 \mu_B$ per formula unit. One of the best experimental values for the saturation
 11 magnetization in SFMO is $\approx 3.7 \mu_B$,⁴ discrepancy attributed to some degree of
 12 disorder between Fe and Mo atoms.⁴⁻⁷

13 In half-metallic materials, the challenge is to obtain higher values of T_C in order
 14 to use these materials for spintronic devices at room-temperature. Among them, the
 15 compounds $\text{Sr}_2\text{FeReO}_6$ and $\text{Sr}_2\text{FeMoO}_6$ have a fairly high T_C around 450 K,^{1,8,9}
 16 the $\text{Sr}_2\text{CrReO}_6$ compound has a higher Curie temperature of 635 K.¹⁰ Experimen-
 17 tal results on SFMO have shown that a substantial increase of T_C can be observed
 18 by increasing the number of conduction electrons resulting from the substitution of
 19 divalent Sr^{2+} by trivalent La^{3+} in the $\text{Sr}_2\text{FeMoO}_6$ system.^{11,12} Associated to this
 20 increase of T_C with the introduction of La^{3+} an important amount of disorder is
 21 induced on Fe and Mo sites, which reduce the magnetization of the SFMO sys-
 22 tem. However, recent results show that the La-doped system becomes magnetically
 23 unstable when it is over doped, giving rise to a decrease of the Curie temperature.¹³

24 The valence and conduction electronic bands of SFMO involve d states of the
 25 Fe and Mo transition metals with nominal configurations Fe^{3+} ($3d^5 : S = 5/2$) and
 26 Mo^{5+} ($4d^1$) with one conduction electron per Mo atom, these bands are slightly
 27 hybridized with oxygen p states lying in between Fe and Mo ions. The localized
 28 d states in Fe and Mo emphasize the strong electron correlation in SFMO, which
 29 increases the T_C .^{14,15} Furthermore, half-metallicity in this compound has been
 30 described within a strong electron–electron interaction between Fe localized elec-
 31 trons and Mo conduction electrons,^{14,16} consistent with experimental results of
 32 photoemission¹⁷ for the ferromagnetic ground state in SFMO.

33 In order to have a full control of the disorder effects in SFMO, some efforts have
 34 been undertaken, studying the Fe-deficient regime and the Fe-rich composition,^{5,7}
 35 the substitution of Sr with a higher cation, such as La,¹¹ or the doping with a smaller
 36 cation, Nd^{3+} for example Refs. 11, 18–20. Nd doping, as the case of La doping,
 37 provides an efficient way to increase the T_C but also decreases the saturation mag-
 38 netization.¹⁹ However, Nd doping could isolate the effect of electron doping in the
 39 conduction band.²¹ In the following, the substitution of Sr^{2+} by Nd^{3+} in the SFMO
 40 double perovskite is analyzed experimentally taking into account the charge neu-
 41 trality, we will consider the system $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ ($y = 0.0, 0.25, 0.5, 0.75$
 42 and 1.0) for the stoichiometric case ($x > 0$ with $x = y/2$) and the off-stoichiometric
 43 one ($x = 0$) and discuss structural and magnetic characterization by different

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1 techniques. However, it is important to emphasize that Nd doping induces disorder
2 on Fe and Mo sites¹¹ provoking the migration of Fe atoms to Mo sites in order to
3 balance the charge introduced by Nd into the cell.

2. Experimental Details

4 Polycrystalline samples of the $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ (SNFMO) compound with
5 $y = 0.0, 0.25, 0.5, 0.75$ and 1.0 for the stoichiometric case ($x > 0$ with $x = y/2$)
6 and the off-stoichiometric one ($x = 0$) were synthesized by the standard solid-state
7 reaction method. The reactants used and provided by Sigma–Aldrich were stron-
8 tium carbonate SrCO_3 (99.9%), neodymium oxide Nd_2O_3 (99.9%), iron oxide Fe_2O_3
9 (99.98%) and molybdenum oxide MoO_3 (99.99%). All the reactants were weighted
10 according to stoichiometric amounts. The oxides and the carbonate were thor-
11 oughly mixed by mechanical milling during 4.5 h forming the first precursor
12 phase $\text{SrMo}_{1-x}\text{O}_4$ and part of the carbonates released in the form of CO_2 . In
13 order to eliminate the remaining carbonates and to obtain the second precursor
14 $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{O}_3$, the mixed powders were calcined at 900°C during 3 h in an
15 oven with a heating ramp of $30^\circ\text{C}/\text{min}$. In addition, powders were synthesized at
16 1200°C under a controlled atmosphere of 1–3% of H_2/Ar followed by a 3 h isotherm
17 to finally obtain the double perovskite SNFMO. Structural characterization was
18 performed by X-ray diffraction (XRD) using a Bruker X-ray diffractometer with
19 $\text{CuK}\alpha$ radiation.
20

21 The oxidation states of Fe and Mo ions were studied by X-ray photoelectron
22 spectroscopy experiments using a Physical Electronics VERSAPROBE II XPS.
23 Moreover, the morphology of the double perovskite compounds was characterized
24 by using a JEOL (JSM-IT300) scanning electron microscope (SEM) under high
25 vacuum (10^{-4} Pa) conditions, the chemical homogeneity and the elemental compo-
26 sition of the compounds were performed by X-ray energy dispersive spectroscopy
27 (EDS) using a 20 mm^2 Oxford X-MaxN Silicon Drift Detector (SDD) coupled to
28 the microscope. The Curie temperature and the hysteresis loops have been obtained
29 using a physical property measurement system (PPMS) from Quantum Design by
30 applying the magnetic field in the range $\pm 1.8\text{ T}$.

3. Results and Discussion

31 The XRD patterns for the stoichiometric case are presented in Fig. 1(a) for
32 different compositions of the double perovskite $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ ($y =$
33 $0.0, 0.25, 0.5, 0.75$ and 1.0) for $x > 0$ (with $x = y/2$). The ordered case SFMO
34 ($y = 0$ and $x = 0$) is also included as a reference pattern, it shows the character-
35 istic reflections reported early.⁹ The average crystallite size of SNFMO is in the
36 range of 30–40 nm. The patterns for $y < 0.5$ ($x = y/2$) show a displacement of the
37 peaks to the left indicating an increase of the lattice parameters and for $y > 0.5$
38 ($x = y/2$) the displacement of the peaks is to the right indicating a decrease of the
39 lattice parameters. The diffraction patterns of the stoichiometric phase for $y = 1$
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1 ($x = 1/2$) show a reflection at approximately 23° corresponding to the NdFeO_3
 2 precursor phase.

3 In order to satisfy the charge neutrality in our Nd doping system, Fe ions tend
 4 to replace partially the sites occupied by Mo ions and the system becomes stoi-
 5 chometric $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ for $x > 0$ (with $x = y/2$), where the number
 6 of itinerant electrons considering Fe (3+) is given by $n = 1 + y - 3x$. For $x = y/2$
 7 ($x > 0$), the itinerant electrons decreases with Nd doping as $n = 1 - y/2$. How-
 8 ever, it is interesting to note some important cases in our discussion; for $x = 0$
 9 (the off-stoichiometric series $\text{Sr}_{2-y}\text{Nd}_y\text{FeMoO}_6$ ^{18,19}), the number of itinerant elec-
 10 trons ($n = 1 + y$) increases with the doping, while for $y = 0$, but $x \neq 0$, the

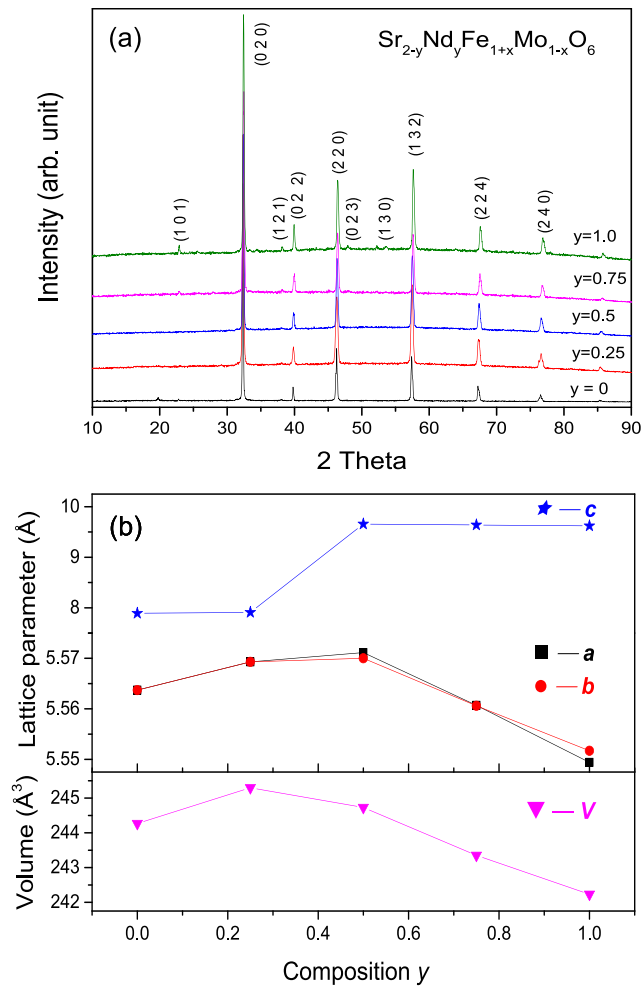


Fig. 1. (Color online) (a) XRD patterns for the double perovskite $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ system with $y = 0.25, 0.5, 0.75$ and 1.0 for $x > 0$ (with $x = y/2$) and (b) lattices parameters and the unit cell volume for all compositions obtained using the Rietveld method.

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1 other stoichiometric series $\text{Sr}_2\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ is obtained,^{5,7} where the number of
 2 itinerant electrons ($n = 1 - 3x$)⁷ decreases for $0 < x < 1/3$ and increases for
 3 $-1 < x < 0$. By observing the limit cases of the latter system, we have that for
 4 $x = -1$ the number of itinerant electrons is $n = 2$ for SrMoO_4 where the valence
 5 of Mo is 4+, and for $x = 1/3$ ($n = 0$) the oxidation state of Mo is 6+, the valence
 6 of Mo cannot exceed 6+ for $x > 1/3$ and Fe cannot conserve 3+. In the limit with
 7 $x = 1$, the valence of Fe is 3+ corresponding to the $\text{SrFeO}_{3-\delta}$ and for $x = 0$ the
 8 valence is 3+ in the $\text{Sr}_2\text{FeMoO}_6$ compound.⁵

9 The crystal structure of the stoichiometric case has been characterized by the
 10 Rietveld refinement technique. The system was indexed for concentrations with $y <$
 11 0.5 using a double perovskite crystal structure for the tetragonal space group $I4/m$
 12 symmetry. However, for $y \geq 0.5$, it was necessary to use the monoclinic space group
 13 $P2_1/m$ to index the crystal structure, which shows a structural transition at $y = 0.5$
 14 composition. This structural transition could be attributed to the smaller ionic size
 15 of Nd^{3+} with respect to Sr^{2+} that induces the rotation of the two-oxygen octahedral
 16 FeO_6 and MoO_6 together with the increases of the (Fe, Mo)-bond due to the electron
 17 filling. In Fig. 1(b), our refinement results show that for the tetragonal structure
 18 the lattice parameters a , b and c increase, while for the monoclinic structure, the
 19 lattice parameters a , b and c decrease. For the transition composition $y = 0.5$,
 20 even though the three lattice parameters increase, the unit cell volume decreases,
 21 see Table 1, because the structural transition involves an important change at the
 22 angle $\beta = 125.18$ between the a and c axis.

23 In Fig. 2, results of XPS for Fe (a) and Mo (b) in the stoichiometric double
 24 perovskite SNFMO system are presented. The spectra for $y = 0$ ($x = 0$) are included
 25 only as a reference. Figure 2(a) shows that the Fe 2p spectrum consists of Fe
 26 $2p_{3/2}$ and Fe $2p_{1/2}$ excitations at 708.4 and 721.8 eV, respectively, indicating that
 27 Fe cations are mainly Fe^{3+} , the spin-energy separations of the two excitations is
 28 13.4 eV; it is important to note that no shift of the peaks is observed, showing
 29 that there is almost no doping effect on Fe valence and the oxidation state 3+
 30 remaining unchanged. Figure 2(b) shows the Mo 3d spectrum consisting of two
 31 peaks from Mo $3d_{5/2}$ and Mo $3d_{3/2}$, excitations which are located at 230.9 and
 32 234.0 eV, respectively. The intensity of these peaks increases as y increases, showing
 33 a shifting from Mo^{5+} to Mo^{4+} with Nd doping. The spin-energy separation of the

Table 1. Lattice parameters and the unit cell volume of the double perovskite $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ compound for $x > 0$ (with $x = y/2$).

Doping (y)	Space group	$a(\text{\AA})$	$b(\text{\AA})$	$c(\text{\AA})$	Vol. ($\text{\AA})^3$
0.0	$I4/m$	5.5637	5.5637	7.8910	244.270
0.25	$I4/m$	5.5693	5.5693	7.9083	245.300
0.50	$P2_1/m$	5.5711	5.5700	9.6563	244.730
0.75	$P2_1/m$	5.5607	5.5606	9.6386	243.353
1.0	$P2_1/m$	5.5494	5.5517	9.6202	242.232

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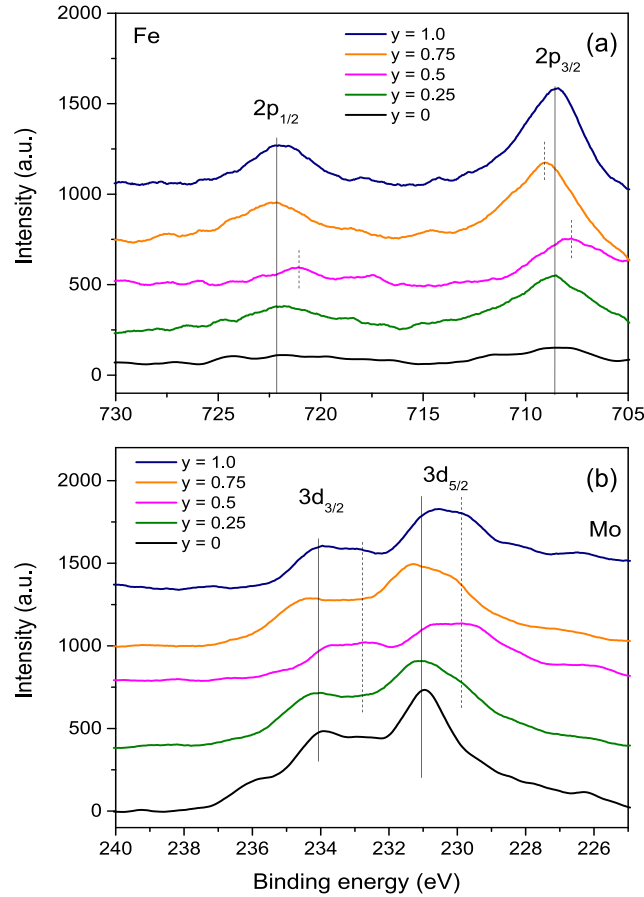


Fig. 2. (Color online) X-ray photoelectron spectra (XPS) for Fe and Mo in the double perovskite $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ compound for $x > 0$ (with $x = y/2$). For Fe (a) the solid lines correspond to Fe^{3+} and for Mo (b) the solid lines correspond to Mo^{5+} and the dashed lines correspond to Mo^{4+} .

1 two main excitations in Mo is 3.1 eV. The valence behavior of Mo is in agreement
2 with previous results for Nd-doping.¹⁹

3 Figure 3 shows the SEM images for different compositions of $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}$
4 $\text{Mo}_{1-x}\text{O}_6$ for $x > 0$ (with $x = y/2$); (a) $y = 0.0$ ($x = 0.0$) is presented only as a
5 reference, (b) $y = 0.25$ ($x = 0.125$) and (c) $y = 1.0$ ($x = 0.5$). In this figure, it is
6 observed that the increase of Nd content leads to larger grains whose boundaries
7 cannot be appreciated, Nd may act as a nucleation center.²² The EDS spectra for
8 $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ are shown in Fig. 4, the EDS microanalysis has been done
9 in different surface regions of each sample and shows the metallic content through
10 a chemical mapping as well as an homogeneous composition of the samples. The
11 Sr/Nd ratio changes according to the stoichiometry. The atomic percentages of each
12 of the elements are shown in Table 2.

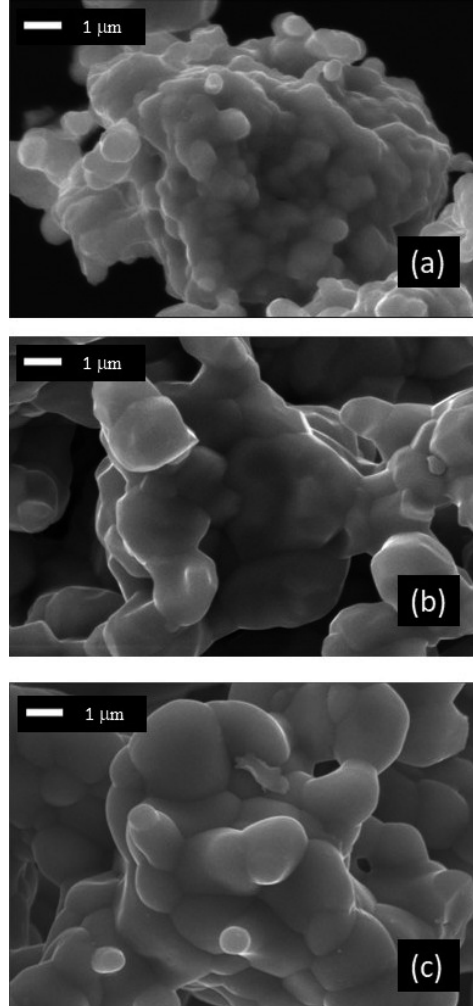
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Fig. 3. SEM images for different compositions of the double perovskite $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ for $x > 0$ (with $x = y/2$), (a) $y = 0.0$ is the reference case, (b) $y = 0.25$ and (c) $y = 1.0$.

1 Susceptibility measurements of the double perovskite SNFMO compound are
 2 presented in Fig. 5. The strong influence of Nd doping can be observed in both
 3 cases for $x = 0$ (off-stoichiometry) and for $x > 0$ with $x = y/2$ (stoichiometry).
 4 For $x = 0$, the Curie temperature decreases at $y = 0.25$, which could be due to the
 5 increment of the structural distortions that prevail over the electronic effects; for
 6 high Nd doping ($y = 0.5$), the compound achieves a higher T_C in good agreement
 7 with $n = 1 + y$. It is important to note that we observe a good agreement with
 8 previous results.²¹ On the other hand, for $x > 0$ ($x = y/2$), a monotonous increase
 9 of T_C is observed even for small y values, this behavior could be associated to
 10 the fact that Nd ions tend to substitute the strontium ions surrounded by iron

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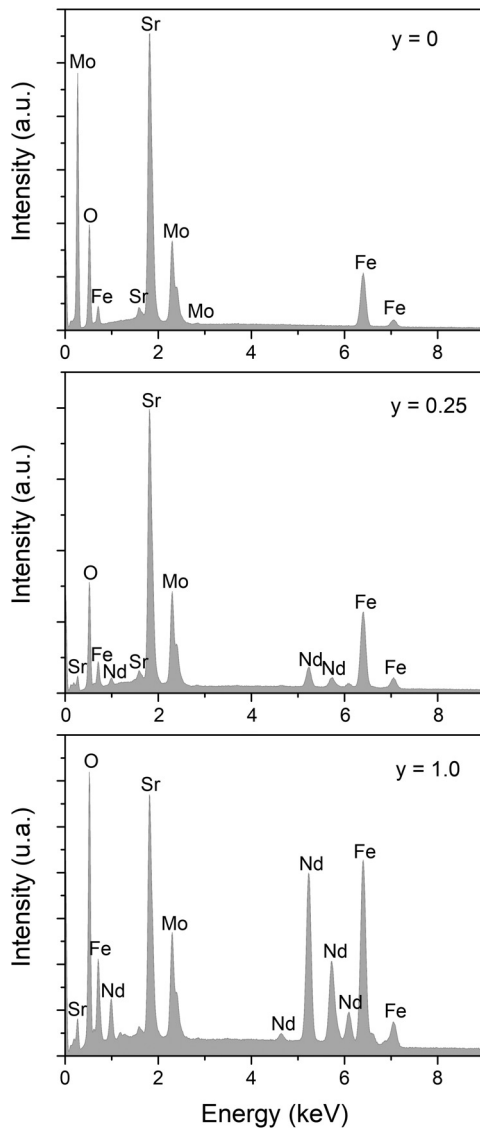


Fig. 4. EDS spectrums for different compositions of the system $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ ($y = 0.0$ as the reference case), $y = 0.25$ and $y = 1.0$ for $x > 0$ (with $x = y/2$).

1 atoms because of their oxidation states. In this case, the structural distortion is
 2 able to overcome the electronic effects. It should be noted that this behavior is
 3 valid for small concentrations of Nd doping since there is a competition with the
 4 Fe enrichment, higher values of doping ($y > 0.5$) do not show T_C .

5 For the magnetic characterization of the two series of $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$
 6 compounds with $x = 0$ and $x > 0$, hysteresis loops have been recorded by
 7 applying the external magnetic field in the range ± 1.8 T, as shown in Figs. 6(a)

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Table 2. Atomic percentages for each of the elements in the $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ compound obtained by EDX microanalysis, $y = 0$ ($x = 0$) is included as a reference.

Element	$y = 0$	$y = 0.25$	$y = 0.5$	$y = 0.75$	$y = 1.0$
O	70.08	64.66	63.33	63.41	62.74
Fe	9.29	12.50	13.22	14.25	15.82
Sr	13.92	13.49	12.30	9.61	7.70
Mo	6.71	7.13	6.44	5.4	4.1
Nd	0	2.23	4.71	7.34	9.64

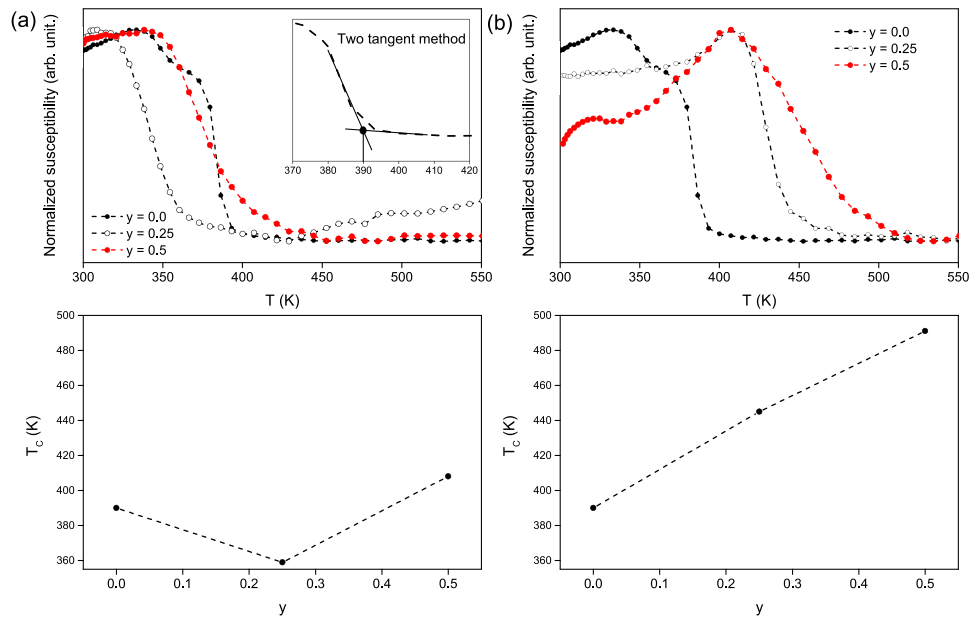


Fig. 5. (Color online) Susceptibility measurements and the Curie temperature for different compositions of the double perovskite system $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ ($y = 0.0, y = 0.25$ and $y = 0.5$). The off-stoichiometric case ($x = 0$) is shown in (a) and the stoichiometric case ($x > 0$, with $x = y/2$) is shown in (b).

1 and 6(b), respectively. The hysteresis loops for both types of compounds show a
 2 magnetization loss of about 90%, showing that replacing half of the sites occupied
 3 by Sr ions leads to an almost nonmagnetic compound. The close views of these
 4 curves at low magnetic field values (see the insets) allow to observe fine details
 5 regarding magnetic parameters like the remanence magnetization and the coercive
 6 field. Clear differences in these parameters between both series of samples
 7 arise when the lack of charge neutrality, due to the substitution of Nd in place
 8 of Sr, is overcome by balancing the Fe and Mo ions in the stoichiometric com-
 9 pounds $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$. Such ionic balance has a significant impact on the
 10 magnetic behavior of the Nd-doped double perovskite, so the loss of ferromagnetism

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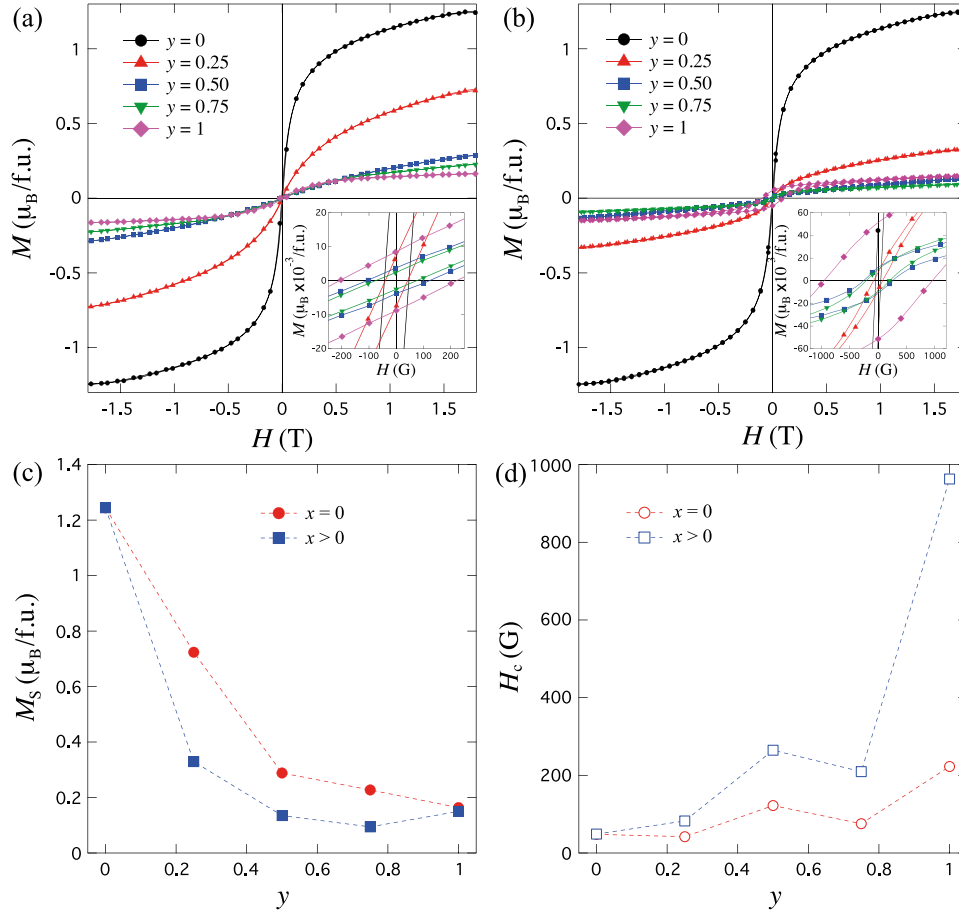


Fig. 6. (Color online) Hysteresis loops recorded in the range ± 1.8 T for double perovskite compounds (a) $\text{Sr}_{2-y}\text{Nd}_y\text{FeMoO}_6$ and (b) $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ with different Nd contents y and for $x > 0$ (with $x = y/2$). Variation with the Nd content of (c) the saturation magnetization (M_s , at 1.8 T) and (d) the coercive field (H_c) for the double perovskite compounds of (a) and (b).

- 1 takes place faster than in the non-ion-balanced compounds, as seen in Fig. 6(c).
- 2 As suggested by a previous work, the progressive reduction of M_s with y for the
- 3 non-ion-balanced compounds can be ascribed to the contribution of the anti-site
- 4 disorder, whereas the further reduction of M_s observed for the ion-balanced com-
- 5 pounds can be ascribed to the electron doping.¹¹ In both cases, with and without
- 6 charge neutrality, the coercive field increases with the Nd content because of the
- 7 larger amount of domain wall pinning sites that delay the magnetization reversal
- 8 process [see Fig. 6(d)]. Moreover, the increase of the coercive field for compounds
- 9 with charge neutrality is consistent with a larger amount of anti-site defects that
- 10 lead to antiferromagnetic regions as in the case of compounds without charge neu-
- 11 trality. However, the further increase for $y > 0.5$ for the former is consistent with

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1 the structural transition observed in Fig. 1(b), which can be responsible for an
2 additional magneto-crystalline anisotropy. Indeed, the modification of the double
3 perovskite unit cell can influence the spin-orbit coupling that is at the origin of
4 such kind of magnetic anisotropy, as suggested elsewhere.²³

5 In conclusion, we have presented an analysis of the structural and magnetic prop-
6 erties of the double perovskite $\text{Sr}_{2-y}\text{Nd}_y\text{Fe}_{1+x}\text{Mo}_{1-x}\text{O}_6$ system. Our results reveal
7 a structural transition, from tetragonal to monoclinic, that occurs for the composi-
8 tion $y = 0.5$. The value of the maximum saturation magnetization is $\approx 1.2 \mu_B/\text{f.u.}$
9 for $y = 0$, well below the optimum theoretical value $4 \mu_B$ due to the large amount
10 of Fe/Mo disorder in the structure. The magnetization decreases as the Nd dop-
11 ing increases, this is mainly caused by the antiferromagnetic interaction between
12 the Fe–O–Fe. The XPS results for the stoichiometry compound show that as the
13 doping increases, the Mo oxidation state shifts from 5+ to 4+ while Fe valence
14 remains constant; this behavior could be associated to the fact that Nd ions tend
15 to substitute the Sr ions surrounded by Fe atoms because of their oxidation states,
16 indicating that electron injection preferentially affects the Mo cations. The Curie
17 temperature increases for all values of Nd doping as soon as the charge neutrality
18 is considered.

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23

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