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Antisites and electron-doping effects on the magnetic transition of Sr_2FeMoO_6 double perovskite

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The effect of antisite (AS) defects and electron doping on the ferromagnetic–paramagnetic (FM-PM) transition of Sr_2FeMoO_6 (SFMO) double perovskite has been studied. From a detailed analysis of the magnetization curves across the FM-PM transition we conclude that AS defects decrease the average strength of the magnetic interactions, although, remarkably enough, some of them are actually enhanced. It follows that the mean-field Curie temperature lowers whereas the onset of magnetization occurs at somewhat higher temperatures. The FM-PM transition has also been analyzed in electron-doped $La_xSr_{2-x}FeMoO_6$, where the AS defects concentration has been found to increase upon La doping. It turns out that in spite of the presence of AS, the mean-field Curie temperature significantly rises upon La and electron doping. This experimental finding contrasts with some recent predictions and emphasizes the role of itinerant electrons in the ferromagnetic coupling in these oxides. Moreover, our results indicate that disordered (i.e., with AS) double perovskite materials should be described as systems with random magnetic anisotropy.

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INTRODUCTION

Double perovskites $A_2BB'O_6$ ($A=$Sr, Ba, Ca; $B=$Fe, Cr, ...; $B'=$Mo, Re, ...) have recently been proposed as good candidates to be used in spin electronics due to their half metallic ferromagnetism and their Curie temperature (T_C) above room temperature[1]. In the ideal Sr_2FeMoO_6 compound, strong ferromagnetic correlations exist between the localized moments of the Fe ions occupying the B sublattice and the conduction band has been predicted to be fully spin (minoritary spins) polarized.

The most studied ferromagnetic double perovskite, Sr_2FeMoO_6 (SFMO), is expected to have an ideal saturation magnetic moment of about $4\mu_B$; however values beyond $3.8\mu_B$ have not been published yet. It is well known that even pure samples present a reduced magnetic moment that strongly depends on the synthesis process.[2] This difference between ideal and actual materials is believed to have its main origin in the lack of cationic ordering in the BB' sublattices. Each cation B that is misplaced in a B' position is called an antisite (AS) defect. From the effects of AS defects on the saturation magnetization[3,4], it can be inferred that since the sublattices B and B' are antiferromagnetically arranged, each antisite contributes to a certain decrease of the saturation moment of the sample.

Whereas the effect of AS defects on the saturation magnetization of the oxide is well understood, the consequences of cationic disorder on the Curie temperature have not been experimentally studied in detail. In spite of this, theoretical models have been elaborated that attribute to AS defects a fundamental role either on the stabilization of the ferromagnetic phase[5] or, conversely, to the suppression of the ferromagnetic order.[3,6] For instance, early Monte Carlo simulations assuming a network of superexchagelike interactions indicated that T_C decreases monotonically with order defects.[3] More recent simulations using a three band model Hamiltonian[6] lead to a nonmonotonic dependence of T_C with AS defects, where T_C was predicted to increase with the first 10% of antisites. Subsequent rising of the AS concentration lowers T_C. The basis behind the initial rising of T_C is that ferromagnetic alignment of next–near–neighboring Fe ions is favored if the intermediate nonmagnetic Mo ion is substituted by an Fe ion. The Fe nearest neighbors couple strongly antiferromagnetically due superexchange, which in turn, strengthens the ferromagnetic coupling of the Fe next–nearest neighbors. In fact, the same idea has been pushed forward by Solovyev[5] who claimed that ferrimagnetic order could not be stabilized by purely electronic mechanisms, but some amount of antisite disorder could do so. It is thus clear that experiments are required to solve this controverted issue for the understanding of the ferromagnetic order in these oxides.

On the other hand, some recent experiments have shown that electron doping—achieved via a partial substitution of divalent Sr^{2+} by trivalent La^{3+} ions in Sr_2FeMoO_6 —may induce a substantial rising of the Curie temperature. In fact, in the system $La_xSr_{2-x}FeMoO_6$ (Ref. 7) it has been claimed that T_C can rise as much as 80 K. This substitution adds electrons to the B-B' sublattices and an increase of the electron density at Fermi level has been observed.[8] We note that this La doping has the detrimental consequence of a reduction of the magnetization due to the increasingly large disorder induced in the Fe/Mo sublattices. However, the experimentally observed enhancement of T_C contrasts with some recent theoretical predictions.[6,9] Indeed, calculations done assuming a rigid band model and neglecting Coulomb electronic correlations in the 4d Mo orbitals, led to the conclusion that electron doping in Sr_2FeMoO_6 should not increase T_C but rather reduce it. We should mention that these discrepancies also become apparent in the study of the evolution of the spin stiffness with La substitution. For example, Moritomo et al.[10] have reported that in $La_xSr_{2-x}FeMoO_6$ (x in the 0–0.3 range) the spin stiffness coefficient (proportional to the interaction strength J) obtained from the $T^{3/2}$ temperature dependence of the magnetization, increases with electron doping,[10] in contrast with theoretical simulations.[11] A possible reason for the discrepancy could be the fact that in $La_xSr_{2-x}FeMoO_6$, as indicated above, La doping is accompanied by an increase of the concentration of AS defects which causes a broadening of the paramagnetic-ferromagnetic (PM-FM) transition and consequently, the determination of the Curie temperature may not be free from uncertainty. Therefore, again it is clear that accurate determination of the Curie temperature in electron doped materials, taking into account the possible contribution of the AS defects, is required.

With the aim to solve these issues, in this work we present a detailed analysis of the effects of the antisites and electron doping on the ferromagnetic–paramagnetic transition of Sr_2FeMoO_6 and $La_xSr_{2-x}FeMoO_6$ double perovskites. Accurate analysis of the magnetization curves $M(T)$ across the PM-FM transition have been performed and the Curie temperature has been determined using different methods: (i) extrapolation of the transition curve to zero magnetization, (ii) inflection point determination via the derivative dM/dT, and (iii) Arrott plots. We will show that the values of T_C and their evolution with antisites substantially depend on the method used for evaluation. We conclude that AS defects decrease the average strength of the magnetic interactions although remarkably enough, some of them are actually enhanced. It follows that, as a result of the presence of AS defects, the FM-PM transition broadens, the mean-field Curie temperature lowers whereas the onset of magnetization occurs at somewhat higher temperature. However, in $La_xSr_{2-x}FeMoO_6$, T_C increases with lantanum substitution independently of the method utilized to determine it. This observation indicates that electron doping, induced by La doping, despite the concomitant structural distortion has the effect of rising the Curie temperature. Thus, the detrimental effect of the induced AS defects is clearly overcomed by reinforced magnetic interactions. Moreover, Arrott plots of the most disordered samples indicate that these systems (pure and electron doped) behave as random anisotropic magnets.

EXPERIMENT

Pure Sr_2FeMoO_6 samples with different amounts of antisites defects and La substituted $La_xSr_{2-x}FeMoO_6$ ($x=0.2$, 0.4, and 0.8) samples have been synthesized using processes described elsewhere[2,7]. Magnetic measurements have been performed using a superconducting quantum interference device and a vibrating sample magnetometer (VSM). The structural characterization of the samples has been carried out using x–ray diffraction. The microstructural parameters, including antisites, were obtained from Rietveld refinements using the FULLPROF (Ref. 12) software. The AS concentration is defined as the ratio of the concentration of B cations in B' positions divided by the total concentration of B cations. Therefore, AS=50% corresponds to a totally disordered double perovskite. Four different Sr_2FeMoO_6 samples have been prepared and measured in this paper; the AS defect concentrations were estimated to be 2.5%, 14%, 25%, and 35%, respectively. In the $La_xSr_{2-x}FeMoO_6$ samples, the corresponding AS values were 11% ($x=0.2$), 34% ($x=0.4$) and 41% ($x=0.8$).

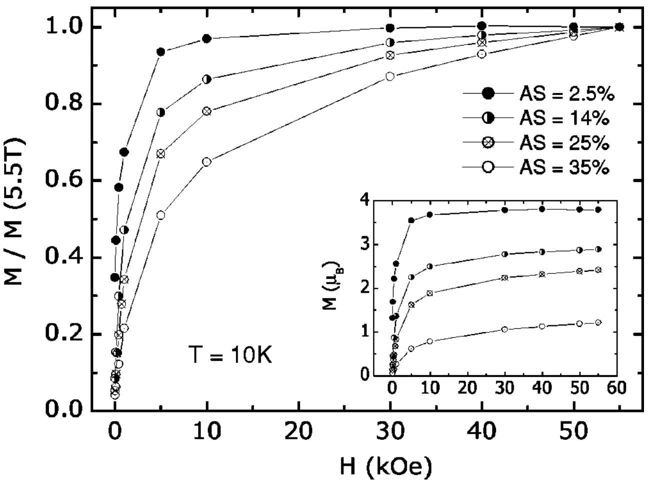

FIG. 1. Dependence of the normalized magnetization, $M/M(5.5T)$, at $T=10$ K for SFMO samples having different antisite (AS) concentrations. Shown in the inset are the magnetization curves.

RESULTS AND DISCUSSION

The saturation magnetization of the pure Sr_2FeMoO_6 samples is found to decrease with the increase of AS, $M_S(H=5.5\text{ T};\ T=10\text{ K})=3.8\mu_B$ (2.5%), $2.9\mu_B$ (14%), $2.4\mu_B$ (25%), and $1.2\mu_B$ (35%)[2] (see the inset in Fig. 1). Shown in Fig. 1 (main panel) is the magnetization versus field normalized to the $M(H=55\text{ kOe})$ values. Data plotted in this way show that AS defects not only reduce, as expected, the saturation magnetization but also promote a magnetic hardening of the system. In other words, it becomes increasingly difficult to saturate the samples as the amount of AS increases.

Differences between the samples with different amounts of antisites also become evident at the PM–FM transition. In Fig. 2 we show the temperature dependence of the normalized magnetization of the most ordered and most disordered SFMO samples, i.e., the Sr_2FeMoO_6 samples having the lowest and highest concentrations of AS (2.5% and 35%, respectively). Inspection of this figure immediately reveals that the FM-PM transition becomes wider as antisites are introduced. Therefore, antisite defects broaden the distribution of magnetic interactions. This can also be clearly observed in the Fig. 2 (inset) where the temperature dependence of the derivatives of the magnetization (dM/dT) of the same samples is shown.

In the most disordered sample a magnetic background extending to high temperature, can be observed. The presence of traces of ferromagnetism at high temperature is quite common in nominally pure Sr_2FeMoO_6 oxide, and it is believed

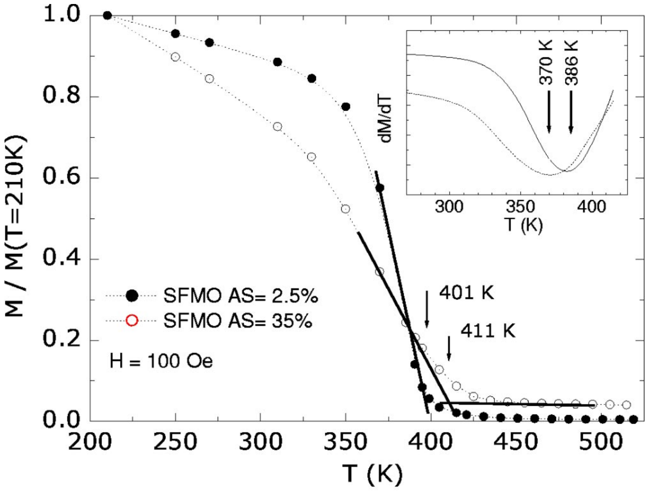

FIG. 2. Temperature dependence of the normalized magnetization, $M/M(210\ \mathrm{K})$, (at $H=100$ Oe) for SFMO samples having different antisite concentrations. The straight lines indicate the linear extrapolation used to determine the extrapolated T_C (indicated by the arrows). In the inset the derivative of the magnetization, dM/dT, of the same sample. The inflection point T_C is indicated by arrows.

that it could originate from tiny amounts of the Fe impurity phase[4,13] that can be hardly avoided in the synthesis of these materials, and it is certainly below the detection limit of x-ray diffraction. Although this spurious signal is only about 10^{-2} emu/g, it becomes apparent in Fig. 2 due to the normalization. It is worth to stress that such small amount of impurity does not have any effect on the determination of T_C by any of the methods to be employed in this work.

In order to analyze the effect of AS defects on the Curie temperature different methods to determine T_C have been used: (i) a linear extrapolation of $M(T)$ to zero magnetization, (ii) a determination of inflexion point of the transition by using the numerical derivative dM/dT, and (iii) Arrott plots. The first two methods are illustrated by data in Fig. 2. The Arrott plots for the same samples are included in Fig. 3 (top and bottom panels). In the Arrott plots, T_C is given by the isotherm displaying a linear M^2 vs H/M variation. In Fig. 3, T_C has been indicated by a straight dashed line. The Curie temperatures extracted by these different methods (i–iii) are collected in Fig. 4. It can be observed that the T_C values obtained by derivative and Arrott plot methods are quite similar and remain roughly constant until a certain amount of antisites defects is reached. Beyond this threshold, T_C decreases steeply. A similar evolution of T_C on AS defects has been reported for in single crystalline specimens, measured with ac susceptibility, where T_C is found to decrease for AS defects beyond 16%.[14] It is important to note that ac susceptibility measurements are essentially zero-field measurements and the results should be free from any field induced effects. Remarkably enough, T_C data extracted by extrapolation clearly show a different dependence on AS defects. The results in Fig. 4 indicate that the extrapolation method gives T_C values that, in the range of AS defects investigated (2.5% –35%), monotonically increase when AS defects increase.

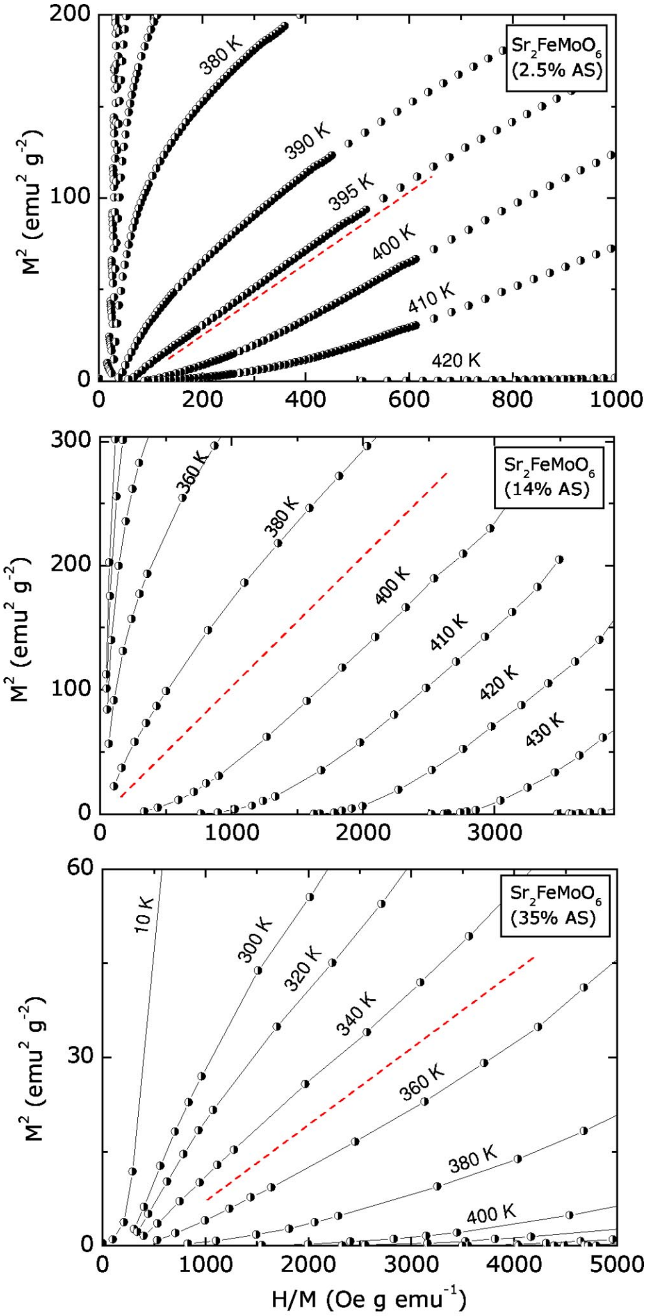

FIG. 3. Arrott plots (mean field) for SFMO samples with different AS concentration: 2.5% (top), 14% (middle) and 35% (bottom). The dashed lines indicate the corresponding T_C isotherms.

These two different evolutions of T_C with antisites reflect that the PM–FM transition gradually broadens. In fact, the T_C values extracted by using these three methods are quite similar for low AS concentrations but become largely different as AS rises. For instance, for AS=35%, the difference between extrapolated and Arrott plot values of T_C is as large as 45 K.

The different evolution of T_C values illustrate that the information provided by the experimental methods used is different: whereas the extrapolated values indicate the onset of spontaneous ferromagnetism and thus reflect the contribu-

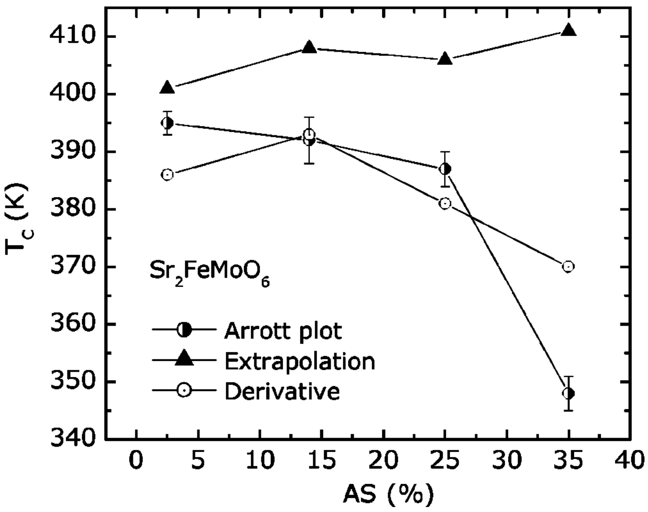

FIG. 4. Dependence of the Curie temperature, using different criteria, on the concentration of antisites for SFMO samples.

tion of the strongest magnetic interactions to the magnetic order. The other two methods provide a measure of the long range ferromagnetic behavior and thus are related to an average intensity of the magnetic interactions.

In summary, antisites in Sr_2FeMoO_6 produce two major effects on magnetic interactions: first, the average strength of these interactions is significantly weakened, and second, some of the magnetic interactions are somewhat reinforced.

At this point it is illustrative to compare in detail the experimental results with some recent models and predictions. Taking into account that in theoretical studies T_C is usually defined as the first temperature at which non–zero magnetization is observed, the simulation results should be better compared to the experimental ones obtained by the extrapolation method, which gives information about the strongest interactions of the distribution. Although Montecarlo simulations[3] predict a monotonic decrease of T_C, Alonso *et al.*[6] found that T_C may increase for AS defects up to 10% of AS defects before it starts to decrease for further increase of the AS fraction. However, the measured extrapolated T_C continuously rises without indications of depression at least up to AS= 35%. This enhancement of some magnetic interactions is compatible with the idea that antisites could maintain ordered the spin of some iron cations over T_C in the neighbors of order defects, as described in the introduction. Evidences of these interactions have been found in Sr_2FeMoO_6 by neutron diffraction.[15]

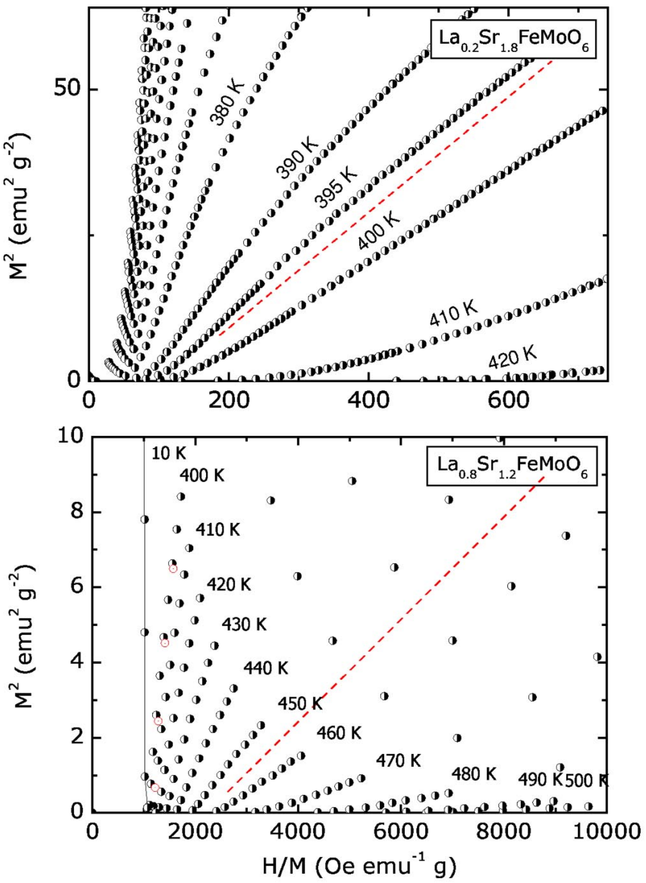

FIG. 5. Arrott plots (mean field) for LSFMO samples with different La concentrations x; $x=0.2$ (top) and $x=0.8$ (bottom).

The role of electron doping has been studied in the $La_xSr_{2-x}FeMoO_6$ series. In this system AS defects are also present and they increase with La substitution almost until total disorder (i.e., AS=50%). It is noteworthy that a substantial increase of the extrapolated T_C with La substitution has been reported[7]. Nevertheless, as we have shown above, in disordered Sr_2FeMoO_6 systems, the evolution of T_C depends on the method used to determine it. Thus, in order to elucidate the role of AS defects and La substitution in the FM-PM transition in the system, T_C has been again analyzed using the three mentioned methods (i–iii): extrapolation, derivative and Arrott Plots. Some of the Arrott plots are shown in Fig. 5 ($x=0.2$ and $x=0.8$), while in Fig. 6 we collect the corresponding Curie temperatures. The main conclusion that can be drawn from these data is that T_C increases with La substitution *independently* of the method used to obtain T_C. For instance, the Arrott plots of these samples show that T_C rises from ~400 to 450 K. Therefore, the La substitution boosts the average strength of the distribution of magnetic interactions. Moreover, since AS defects actually decrease the average strength of the distribution of interactions and

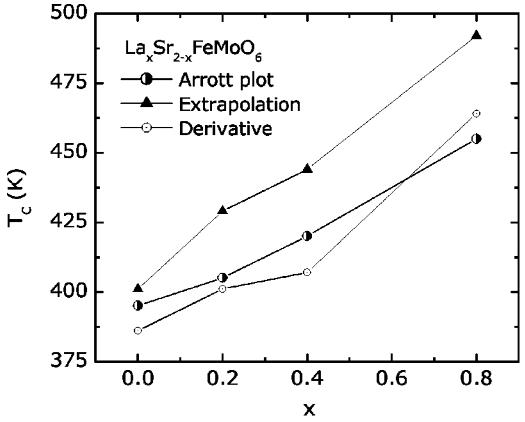

FIG. 6. Dependence of the Curie temperature, using different criteria, on the La concentration x for LSFMO samples.

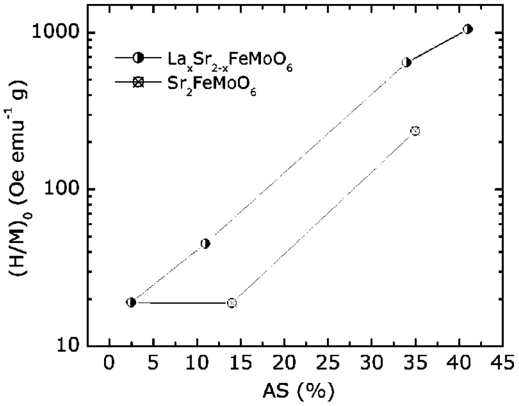

FIG. 7. Dependence of the crossing points of the Arrott plots, $(H/M)_0$ on the concentration of antisites for the SFMO and LSFMO series.

that the AS concentration increases with La substitution it is clear that T_C of electron doped samples without AS should still be higher. Indeed, Serrate *et al.*[16] have been able to synthesize La substituted—and thus electron doped—$(Ba_{0.8}Sr_{0.2})_{2-x}La_xFeMoO_6$, with a negligible concentration of AS defects. In agreement with the discussion presented above it was found that the $d(T_C)/dx = 3$ K/% is substantially larger than in the case of $La_xSr_{2-x}FeMoO_6$ where $d(T_C)/dx = 1.3$ K/%.[7]

At this point one should note that neither in this case, the experimental results match with existing theoretical predictions. Namely, the prediction that electron doping should reduce T_C. The origin of this fundamental discrepancy should be related to some of the hypothesis of the models such as the rigid band electronic configuration and the assumption that electronic correlations in Mo sites are negligible. Indeed, as mentioned in the introduction, although Mo is not magnetic, magnetic correlation could have been induced in its outer bands by polarized itinerant electrons or the Fe–O–Mo hybridization.[17–19] In addition, the presence of La ions substituting Sr is known to promote a structural modification[7,20] that likely produces a modification of the relevant interatomic hopping integrals, which, so far, have not been included in the theoretical models.

Finally, a more in depth analysis of the Arrott plots for both the Sr_2FeMoO_6 and $La_xSr_{2-x}FeMoO_6$ samples reveals some deviations from mean field model predictions, that become progressively more apparent as AS defects increase. In Fig. 3 we showed the Arrott plots corresponding to SFMO samples with different amounts of AS. For the most ordered sample (AS$\approx$2.5%) the plot shows the typical shape of a mean field ferromagnet,[21] with the straight–line isotherm—indicating the Curie temperature—crossing the (H/M) axis at $H/M=0$. This means that at T_C the susceptibility diverges. However, in the Arrott plots of the more disordered samples (Fig. 3, middle and bottom panels) it is clear that the straight-line isotherm does not extrapolate to $H/M=0$ but to a finite value $(H/M)_0$. This produces a shift in the H/M axis, most clearly seen in the 35% disordered sample. This means that at the Curie temperature the susceptibility does not diverge for large amounts of AS defects. Actually, to ensure that the lack of divergence of the susceptibility in the whole temperature range, the 10 K isotherm has been included in Fig. 3 (bottom). In Fig. 7 we collect the shift $(H/M)_0$ as a function of the AS concentration.

The Arrott plots for some $La_xSr_{2-x}FeMoO_6$ samples are depicted in Fig. 5. A similar $(H/M)_0$ shift, although larger, can be clearly appreciated. This observation is fully consistent with the higher AS concentration—and thus larger disorder—in these samples. In Fig. 7 we also include the corresponding $(H/M)_0$ values as a function of AS. It is clear that $(H/M)_0$ increases for both LSFMO and SFMO samples when rising AS defects. We note however, that for LSFMO and SFMO samples of similar AS concentration, $(H/M)_0$ is larger for the former than for the latter; this effect could be due to additional disorder in the magnetic ion environments associated to the random distribution of La/Sr ions and the resulting crystallographic local distortions.

It is noteworthy that such shifts in the Arrott plots, indicating the absence of susceptibility divergence were predicted theoretically by Aharony *et al.* in random field ferromagnets,[22] where the magnetic order is only short ranged. This could be the case of a disordered double perovskite, since antisites produce different magnetic environments, randomly arranged, for the B-B' cat ions, hence the field locally felt by the atoms would be random. The present results may constitute an experimental evidence of the short range magnetic order suggested for disordered double perovskites.[14]

Before concluding, we should mention that Salamon *et al.*[23] reported an analyis of the magnetization curves of a SFMO crystal and determined the critical exponents of the PM-FM transition. The extracted values of the critical exponents for SFMO were $\beta=0.385$ and $\gamma=1.3$,[23] which differ from the mean field ones ($\beta=1/2$, $\gamma=1$) that we used to construct the Arrott plots. The AS concentration in the crystal measured by Salamon *et al.* was not reported, nor the saturation magnetization, so the relevance of the AS defects to the values of the extracted critical exponents cannot be addressed. In the light of the present data, it is clear that AS defects should be properly taken into account when analyzing the details of the magnetic transitions. However, from a practical point of view, no significant differences on the Arrott plots of our samples (Figs. 3 and 5) are observed if data are plotted using the critical exponents of Yanagihara *et al.*[23] instead of the mean-field values we have used. Thus, the extracted Curie temperatures (Figs. 4 and 6) remain almost unchanged.

CONCLUSIONS

Summarizing, the effects of antisite defects and electron doping (La substitution) on the ferromagnetic-paramagnetic transition of Sr_2FeMoO_6 double perovskites have been studied. It has been found that antisites, and the resulting mixing of magnetic interactions, promote a broadening of the PM-FM transition. Whereas the Curie temperature as determined by Arrott plots or the inflecion point criteria inevitably reduces when increasing the AS concentration, this is not so for the onset of magnetization. In fact, for moderate AS concentrations, the onset of magnetization is increased by AS defects. This behavior can be rationalized by the existence of strong magnetic (antiferromagnetic) interactions between misplaced Fe ions at Mo sites and Fe ions at regular sites which contribute to the stabilization of local ferromagnetic coupling between next-nearest-neighbor Fe ions. Due to the presence of some sort of random distribution of magnetic interactions, the magnetic susceptibility does not diverge at the Curie temperature; this behavior is reminiscent of that predicted for random field ferromagnets. In electron doped materials, such as $La_xSr_{2-x}FeMoO_6$, in spite of the same trends associated to the presence of AS defects, the effect of La substitution is so prominent that it overcomes the detrimental contribution of AS defects, and consequentlty, T_C rises with electron doping. Therefore, it is clear that strategies to reduce or eventually suppress the Fe/Mo disorder in electron doped materials should be dealt with in order to obtain materials with opportunities in magnetoelectronics.

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