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Disclosing the origin of the reduced magnetoresistance in electron-doped double perovskites

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Abstract

Electron doping in $A_{2-x}L_x\text{FeMoO}_6$ (where L is a trivalent lanthanide and A is a divalent cation) double perovskites has been established as a suitable technique for increasing their Curie temperature. However, it was found that the magnetoresistance gradually decreases with increasing lanthanide substitution. Here we analyse in detail the magnetoresistance as a function of the magnetic field for several series of ceramic $A_{2-x}L_x\text{FeMoO}_6$ oxides, showing that the data can be well described by assuming a gradual loss of spin polarization of the conduction band upon electron doping. This observation introduces some constraints to models of ferromagnetic coupling in double perovskites.

(Some figures in this article are in colour only in the electronic version)

The growing interest in the development of spintronics devices has stimulated the search for materials with large spin polarization and ferromagnetic order above room temperature. In particular, much effort has been devoted in the past years to the study of ferromagnetic double perovskites of formula $A_2\text{FeMoO}_6$ ($A = \text{Ca}, \text{Sr}, \text{Ba}$). Triggered by the work of Kobayashi *et al* [1], the research has mostly been focused on the study of the compound $\text{Sr}_2\text{FeMoO}_6$ (SFMO), which presents a high Curie temperature ($T_C \sim 400$ K) and displays a strong magnetoresistance (defined as $\text{MR} = (\rho(H) - \rho(0))/\rho(0)$, where $\rho(H)$ is the field dependent resistivity) up to room temperature [1]. The MR in double perovskites is dominated by the so-called tunnelling magnetoresistance (TMR). This effect takes place when conduction electrons tunnel between magnetic domains separated by insulating barriers. As the tunnelling is spin dependent, the transmission probability (and thus the electrical conductivity) is maximized if the moments of the magnetic domains are oriented in a parallel configuration. This alignment can be achieved by the application of a moderate magnetic field. According to Juliere's model [2], the TMR effect is optimized if the conduction band of the ferromagnetic electrodes presents a full spin polarization, that is, if all the carriers show the same spin orientation. Band structure calculations suggest that this is the case for SFMO and related double perovskites [1, 3],

predicting a conduction band with a finite density of states in the spin-down channel and zero density of states in the spin-up channel.

In ceramic double perovskite samples, the insulating barriers associated with the TMR effect are provided by the physical grain boundaries. This is supported by the fact that the strong magnetoresistance observed in ceramic SFMO [1] is not present in specimens where grain boundaries are scarce, such as single crystals [4] or epitaxial thin films [5], and by the fact that it can be very much controlled by modifying the inter-grain interfaces [6, 7]. However, we should mention that the existence of insulating Fe–O–Fe antiferromagnetic patches has been proposed as an alternative origin for the tunnel barriers [8]. As these patches arise from the existence of cationic defects, and are not particularly localized in any region of the sample, this scenario would suggest a weak dependence of the TMR effect on the grain boundaries.

Although the T_C of SFMO is above room temperature, the achievement of double perovskites with even higher magnetic ordering temperatures is desirable for technological reasons. Different strategies for increasing the Curie temperature of double perovskites have been investigated [9, 10]. In particular, electron doping, achieved by means of the substitution of the divalent A cation by a trivalent lanthanide (La^{3+} [9], Nd^{3+} [11]), has been reported as a fruitful approach. The success of this strategy has been extensively checked in different systems such as $\text{La}_x\text{Sr}_{2-x}\text{FeMoO}_6$ [9, 12], $\text{Nd}_x\text{Sr}_{2-x}\text{FeMoO}_6$ [11, 13], $\text{Nd}_x\text{Ca}_{2-x}\text{FeMoO}_6$ [14], $\text{La}_x\text{Ba}_{2-x}\text{FeMoO}_6$ [15], $(\text{Ba}_{0.8}\text{Sr}_{0.2})_{2-x}\text{La}_x\text{FeMoO}_6$ [16], or even in $(\text{Ca, Sr})_{2-x}\text{La}_x\text{CrWO}_6$ [17]. However, it was observed that the increase in T_C was accompanied in all cases by a strong decrease in the magnetoresistance [9, 11, 14, 16], the functional properties of the materials being seriously affected in this way. Two mechanisms have been proposed as being responsible for this undesired effect upon electron doping. In the first place, a loss of spin polarization, originating from the critical increase in cationic disorder upon electron injection [11, 14]. Secondly, the existence of spin-independent inter-granular conduction channels [16], arguably related to chemical modifications on the grain boundaries. Apart from these speculations, the physical reason for the detriment of the magnetoresistance remains unknown.

Here we review the magnetotransport data obtained for different electron-doped ceramic series, namely $\text{La}_x\text{Sr}_{2-x}\text{FeMoO}_6$ (LSFMO; $0 \leq x \leq 0.6$), $\text{Nd}_x\text{Sr}_{2-x}\text{FeMoO}_6$ (NSFMO; $0 \leq x \leq 0.6$) and $\text{Nd}_x\text{Ca}_{2-x}\text{FeMoO}_6$ (NCFMO; $0 \leq x \leq 0.8$). With the aim of shedding light on the origin of the reduction of magnetoresistance upon lanthanide substitution, we have analysed the evolution of the magnetoresistive response of all series by using a model for spin polarized transport in granular media. The proposed model takes into account the dependence of the magnetoresistance on the spin polarization, as well as on the presence of spin-independent inter-granular conduction channels. Our analysis suggests that the spin polarization decreases with electron doping as a consequence of the filling of minority spin states of the conduction band. The implications of this finding will be addressed.

Ceramic samples were synthesized by means of standard solid state reactions, as described elsewhere [18, 11, 14]. A final treatment was performed in a reducing H_2 –Ar (1%) atmosphere, at 1250 °C, leading to micrometric, well-sintered grains. Details of the structural characterization can be found elsewhere [18, 11, 14]. The Curie temperatures, determined by using a variety of techniques such as standard magnetometry or neutron powder diffraction, were found to increase from $T_C \sim 400$ K (SFMO) to 440 K (NSFMO and LSFMO, both with $x = 0.6$) [18], and from $T_C = 385$ K (CFMO) to 450 K (NCFMO, $x = 0.8$) [14]. Photoemission experiments showed that injected electrons indeed increase the density of Mo-related states near the Fermi level [19], reflecting the filling of the conduction band.

Figure 1(a) shows the evolution of antisite (AS) defects—defined as Fe(Mo) ions misplaced in Mo(Fe) positions—as a function of the trivalent substitution for all the series.

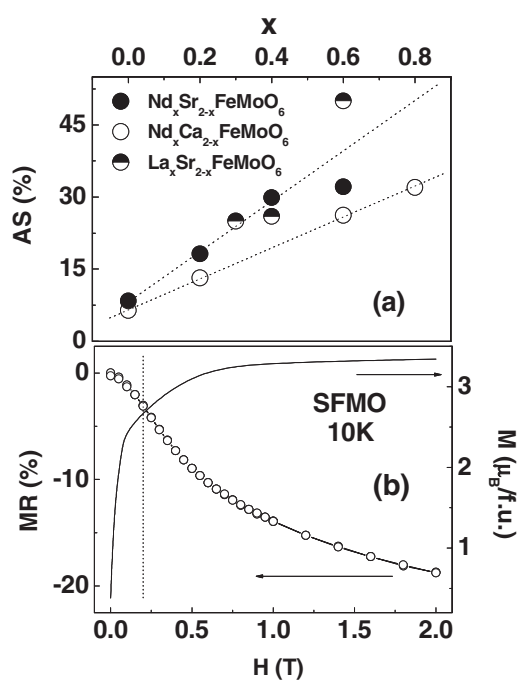


Figure 1. (a) Evolution of the antisite defects—as extracted from the Rietveld refinements of XRD patterns—as a function of doping (x) for $\text{La}_x\text{Sr}_{2-x}\text{FeMoO}_6$, $\text{Nd}_x\text{Sr}_{2-x}\text{FeMoO}_6$ and $\text{Nd}_x\text{Ca}_{2-x}\text{FeMoO}_6$ series. Lines are guides for the eye. (b) Magnetization and magnetoresistance as a function of the applied field for a $\text{Sr}_2\text{FeMoO}_6$ sample. The dotted line at $H = 2$ kOe is intended to mark the rapid saturation of the magnetization against the slower growth of the magnetoresistance.

It can be observed that the cationic disorder increases with electron doping. This behaviour is related to the selective localization of injected electrons on Mo sites [19, 20], which reduces the charge difference between Fe and Mo cations and thus weakens the driving force that controls the Fe–Mo order [10]. The evolution of both the magnetoresistance and the magnetization as a function of the applied field for a SFMO sample (corresponding to the sample with $x = 0$ of the NSFMO series) is shown in figure 1(b). It can be seen that the magnetization shows a sharp increase at relatively low fields, reflecting the rapid orientation of the ferromagnetic domains upon the application of a magnetic field. According to the usual TMR mechanism, as the magnetic field aligns the magnetic moments of neighbouring grains, the MR should also display a concomitant drop, mimicking the M versus H response. This behaviour was well established for other oxide systems such as manganites [21] or CrO_2 [22]. However, inspection of figure 1(b) shows that the MR grows at a much slower rate than the magnetization. For instance, while the ratio $M(H = 2 \text{ kOe})/M(H = 20 \text{ kOe})$ is about 80%, the corresponding $\text{MR}(H = 2 \text{ kOe})/\text{MR}(H = 20 \text{ kOe})$ ratio is below 20%. Additionally, the absence of saturation of the magnetoresistance up to the higher measured field (2 T) is worth noting. This behaviour of SFMO was emphasized by Sarma *et al* [23], who proposed a modified scenario for the usual TMR effect, suggesting a resonant tunnelling mechanism in which it is necessary to magnetize the grain boundaries before achieving a magnetoresistive response [23]. As the grain boundaries are expected to be magnetically harder than the bulk material, the evolution of the magnetoresistance with the applied field should no longer mimic the behaviour of the overall magnetization (largely dominated by the bulk contribution), as observed in figure 1(b).

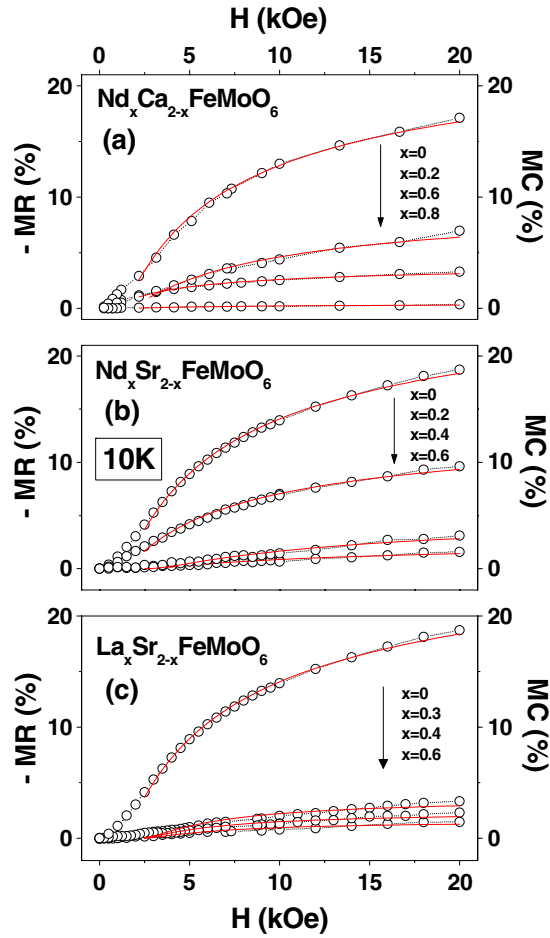


Figure 2. Evolution of the magnetoresistance (MR, left axis) and magnetoconductance (MC, right axis) with applied field for (a) $\text{Nd}_x\text{Ca}_{2-x}\text{FeMoO}_6$, (b) $\text{Nd}_x\text{Sr}_{2-x}\text{FeMoO}_6$ and (c) $\text{La}_x\text{Sr}_{2-x}\text{FeMoO}_6$ series, respectively. Experimental data are shown by symbols, while the fittings—performed by using the model described by equation (5)—are shown by full lines. Data were recorded at 10 K.

Figures 2(a)–(c) show the measured magnetoresistance (left axis) and magnetoconductance ($\text{MC} = (\sigma(H) - \sigma(0))/\sigma(0)$), where $\sigma(H)$ is the field-dependent conductivity (right axis) as a function of the applied field for the studied series. The three series clearly show a systematic decrease of the magnetoresistive response upon electron doping. We will focus on this issue in the following paragraphs.

According to the model developed by Inoue and Maekawa [24] for granular magnetic materials, the magnetoconductance is given by

$$\text{MC} = P^2 m^2 \quad (1)$$

where $P = n_\uparrow - n_\downarrow / n_\uparrow + n_\downarrow$ is the spin polarization of the bulk material and $m = M(H)/M_{\text{Sat}}$ is the reduced magnetization of the grains (n_\uparrow and n_\downarrow correspond to the spin-up and spin-down populations of the conduction band, respectively, while M_{Sat} is the saturation magnetization). According to the previous discussion, in the case of double perovskites the bulk magnetization

appearing in equation (1) should be replaced by the grain boundary magnetization. It has recently been suggested [25] that the magnetic behaviour of the grain boundaries in double perovskites can be described by using a functional form characteristic of spin glasses with a weak anisotropic field [26]

$$m(H) = (1 - a/H^{1/2}) \quad (2)$$

where a is a field-independent parameter. We notice that a similar functional dependence of the magnetization was first used by Dieny *et al* [27] to describe the magnetization of some multilayered ferrimagnetic systems, and subsequently applied by several authors [28] to analyse the role of antiphase boundaries existing in magnetite on the magnetization and the magnetoresistance. Inserting this functional dependence into equation (1), we get

$$MC = P^2(1 - a/H^{1/2})^2. \quad (3)$$

This equation holds for a scenario where inter-grain transport takes place entirely through a spin-dependent channel, and it has already been used to extract the spin polarization of $(\text{Ba}_{0.8}\text{Sr}_{0.2})_2\text{FeMoO}_6$ ceramic samples [25]. We will now take into account the possible existence of an additional spin-independent channel, which could plausibly exist as consequence of slight chemical modifications on the grain boundaries (i.e. oxygen vacancies, or even small amounts of segregated phases). If both channels coexist in a parallel configuration, the total conductivity would be given by

$$\sigma_T = \sigma_{\text{SD}} + \sigma_{\text{SI}} \quad (4)$$

where σ_{SD} and σ_{SI} correspond to the conductivities of the spin-dependent and spin-independent channels, respectively.

Considering both contributions, the magnetoconductance is given by

$$MC = P^2/(1 + \sigma_{\text{SI}}/\sigma_{\text{SD}})(1 - a/H^{1/2})^2 = P_{\text{eff}}^2(1 - a/H^{1/2})^2 \quad (5)$$

where $P_{\text{eff}} = P/(1 + \sigma_{\text{SI}}/\sigma_{\text{SD}})^{1/2}$ is an *effective* polarization. Notice that P_{eff} is bounded between $0 < P_{\text{eff}} < P$, the upper bound ($P_{\text{eff}} \sim P$) being obtained for samples where the transport is dominated by the spin-dependent channel ($\sigma_{\text{SD}} \gg \sigma_{\text{SI}}$) and the lower bound ($P_{\text{eff}} \sim 0$) taking place in the opposite situation ($\sigma_{\text{SD}} \ll \sigma_{\text{SI}}$).

Inspection of the data in figures 2(a)–(c) clearly shows that, for all series, the MC increases with H but decreases upon lanthanide substitution, therefore suggesting that $P_{\text{eff}}(x)$ gradually lowers with x . In order to extract $P_{\text{eff}}(x)$ we have fitted the data in figures 2(d)–(f) using equation (5). The singular nature of the $a/H^{1/2}$ term for zero field precludes using the low field region ($H < H_0$) of the $MC(H)$ curves for fitting. We have found that below 0.25 T the fits critically depart from the experimental data, so we have set $H_0 = 0.25$ T. As evidenced by the solid lines through data in figures 2(d)–(f), experimental data can be well reproduced by equation (5), thus allowing us to extract P_{eff} for each sample.

Figure 3(a) displays the P_{eff} values corresponding to the different series as a function of doping (x). It clearly reflects a decrease in the effective polarization upon trivalent substitution. Inspection of figure 3(a) shows that the maximum P_{eff} value corresponds to both non-doped samples (SFMO and CFMO), and is about 60%. This value is smaller than the $P \approx 85\%$ extracted for SFMO electrodes in magnetic tunnel junctions [29] or granular $(\text{Ba}_{0.8}\text{Sr}_{0.2})_2\text{FeMoO}_6$ samples [25], and suggests the presence of spin-independent conduction channels in our samples, leading to $P_{\text{eff}} < P$.

At this point it is worth noting that the observed evolution of P_{eff} would truly reflect the evolution of the bare spin polarization P only if the ratio $\sigma_{\text{SI}}/\sigma_{\text{SD}}$ remains nearly invariant for the different samples. In order to clarify this, we recall that the presence of spin-independent conduction channels (parameterized in our model by the conductivity σ_{SI}) should be strongly

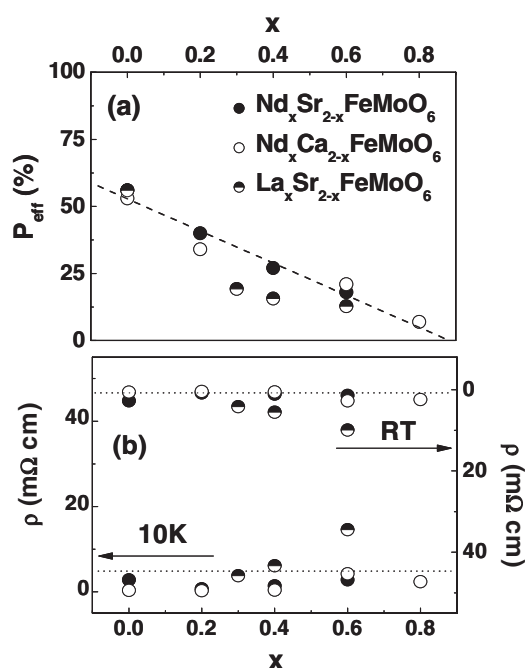


Figure 3. (a) Fitted effective spin-polarization (P_{eff}) as a function of electron doping (x) for $\text{Nd}_x\text{Ca}_{2-x}\text{FeMoO}_6$, $\text{Nd}_x\text{Sr}_{2-x}\text{FeMoO}_6$ and $\text{La}_x\text{Sr}_{2-x}\text{FeMoO}_6$ series; the line is a guide to the eye. (b) Room temperature (RT) and 10 K resistivities corresponding to the same series, the lines are guides to the eye.

dependent on the synthesis conditions [6]. In the first place, we notice that all our samples were synthesized under the same conditions, so it is reasonable to speculate that the $\sigma_{\text{SI}}/\sigma_{\text{SD}}$ ratio cannot vary substantially from sample to sample. On the other hand, we recall that the magnetoresistance of SFMO can be modified by performing controlled grain boundary oxidations [6, 7]; interestingly enough, this process is accompanied by a strong variation of the overall resistivity that can reach several orders of magnitude [6, 7]. Certainly this is not the case for our samples, where the 10 K resistivities (shown in figure 3(b)) of NSMO, LSMO and NCMO series display values between 1–3 m Ω cm, 4–14 m Ω cm and 0.5–4 m Ω cm, respectively. Notice that according to Niebieskikwiat *et al* [6, 7] these slight variations in the resistivity values should not induce important modifications in the magnetoresistive signal. In consequence, we can safely conclude that the grain boundaries of our series do not show substantial variations from sample to sample, indicating that the observed shrinkage of P_{eff} upon lanthanide substitution reflects a true decrease of the real spin polarization P .

We recall that theoretical calculations for ordered SFMO predict that the hybridization of Mo 4d and Fe 3d states (both taking part of the conduction band) induces a strong exchange splitting—parameterized by an energy U —between spin-up and spin-down Mo 4d levels [30]. This leads to a 100% spin polarization of the conduction band, as shown in figure 4(a). In that case, only the spin-down conduction channel would be filled upon electron doping, and the spin polarization would be expected to be conserved. Interestingly enough, the reduction of spin polarization upon electron doping in our samples indicates the filling of *both* spin channels, suggesting a reduced exchange interaction that partially closes the gap between spin-up and

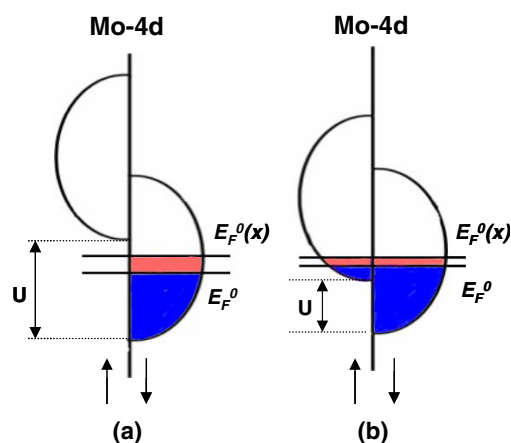


Figure 4. Schemes showing the Mo-4d (t_{2g}) orbitals of $A_2\text{FeMoO}_6$ double perovskites. The splitting exchange energy between both spin channels is indicated as U . Shaded areas point out occupied orbitals. E_F^0 and $E_F(x)$ indicate the Fermi levels of undoped and electron-doped compounds, respectively. (a) Strong exchange situation: the split between both sub-bands is strong enough to allow a full spin polarization, even after electron doping. (b) Moderate exchange situation: the band splitting is weaker than in the latter case, while both spin channels display a non-zero population at the Fermi level. The polarization gets lower upon electron injection.

spin-down Mo 4d states. This would lead to a non-zero density of states at the Fermi level in both spin channels, as schematized in figure 4(b).

It could be argued that the observed increase of antisites upon electron injection (figure 1(a)) can possibly account for a weakening of U as a consequence of the suppression of Fe–Mo–Fe paths [31]. Indeed, it has been observed that the presence of antisites may lower the magnetoresistance [7, 33] of SFMO. However, it is worth noting that the extracted P_{eff} values do not seem to be strongly correlated with the AS concentration. Indeed, comparison of the AS versus x data in figure 1(a) and the P_{eff} versus x data of figure 3(a) clearly illustrates that whereas the $P_{\text{eff}}(x)$ data for all series collapse onto a single line, this is not the case for $\text{AS}(x)$. This observation suggests that upon lanthanide doping, the variation of P_{eff} is dominated by the electron doping rather than by the accompanying antisite defects. The observation of a critical reduction of magnetoresistance in the $(\text{BaSr})_{2-x}\text{La}_x\text{FeMoO}_6$ electron-doped series with much reduced [16, 32] cationic disorder supports the above conclusion. We notice that the extreme sensitivity of P_{eff} to the electron density could naturally be explained if the splitting energy between the spin-down and spin-up sub-bands of the ordered $A_2\text{FeMoO}_6$ ($A = \text{Sr}$ or Ca) double perovskites is weaker than that assumed from band structure calculations, indicating a rather marginal half-metallic character. Indeed, the largest spin polarization measured so far in double perovskites ($\sim 85\%$, according to Bibes *et al* [29]) is smaller than expected (100%). Further experiments such as spin-polarized photoemission are required to definitively settle this question.

In summary, by analysing the magnetotransport data for different electron-doped double perovskites we have enquired about the origin of their reduced magnetoresistance. We have found a critical loss of spin polarization, indicating the filling of the minority spin states of the conduction band. This points to a scenario characterized by a moderate exchange splitting energy of Mo-4d levels, even in the presence of few cationic defects, and suggests a severe drawback to the possibility of obtaining electron-doped double perovskites with increased Curie temperatures *and* high spin polarization.

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