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Effect of cationic disorder on the spin polarization in FeMo double perovskites

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Abstract – Ordered Sr_2FeMoO_6 is expected to have complete spin polarization, however all samples usually present some degree of Fe/Mo disorder which reduces the tunneling magnetoresistance in granular samples. It can be inferred that disorder is detrimental to the half-metallicity. We present an electronic approach of this disorder using a model based on a correlated electron picture with localized Fe-spins and conduction electrons interacting with the local spins via a double-exchange–type mechanism. This succeeds in stabilizing a ferromagnetic ground state in the absence of disorder. Disorder is treated within the dynamical mean-field approach which is equivalent to the coherent potential approximation. We shall show how electron disorder affects the density of states and the ground-state properties with a particular emphasis on the spin polarization.

Half-metallic materials, in which only one-spin direction is present at the Fermi level, are extensively searched for in view of spintronics applications based on magnetoresistance (MR) effects. Among the double-perovskites family $A_2MM'O_6$ (A being a divalent cation and M, M' two transition metals), a lot of attention has focused on the Sr_2FeMoO_6 because of its high Curie temperature $T_C \sim 400\,K$, complete spin polarization and substantial low-field magnetoresistance (LFMR) at room temperature as compared to manganites [1], characteristics highly desirable for magnetoelectronic applications.

In granular materials LFMR is dominated by the intergrain spin-dependent tunneling magnetoresistance (TMR) through insulating grain boundaries (GB), therefore reflecting the spin polarization of the ferromagnetic grains. In the zero-field case, all grains have different magnetization orientations and the intergrain tunneling is reduced due to the strong polarization of the itinerant electrons. Of course the tunneling conductance will be favoured by the alignment of the magnetization of the neighboring grains under application of the magnetic field. According to Inoue and Maekawa [2], the MR (defined as $\mathrm{MR}=\frac{\Delta\rho}{\rho_0}=\frac{\rho(H)-\rho(0)}{\rho(0)}$, $\rho(H)$ being the field-dependent resistance as a function of the applied field H) is given by $\mathrm{MR}=-\frac{P^2m^2}{1+P^2m^2}$, where $P=\frac{\mathcal{N}_\uparrow-\mathcal{N}_\downarrow}{\mathcal{N}_\uparrow+\mathcal{N}_\downarrow}$ represents the spin polarization of the grains, $\mathcal{N}_\uparrow$ and $\mathcal{N}_\downarrow$ are the densities of states at the Fermi level for the up and down electrons in each ferromagnetic grains; $m=\frac{M(H)}{M_S}$ is the relative magnetization of the system, M_S being the saturation magnetization achieved when the magnetizations of all grains are parallel to the magnetic field. Recently, it has been shown that the magnetic state of the GB regions was the relevant ingredient to explain the TMR rather than the bulk magnetization $m(H)$ [3]. A close scenario of "grain boundary magnetism controlled" TMR has been emphasized by Sarma *et al.* [4]. The TMR phenomena is strongly enhanced by a high degree of spin polarization P in the ferromagnetic regime, which can be at most -50% for full polarization $P=\pm 1$, when the saturation magnetization is reached. The half-metallic fully polarized character of ordered Sr_2FeMoO_6 has been

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predicted by band-structure calculations [1]. However, to our knowledge, there are very few direct experimental measures of the spin polarization. $P \approx 0.85$ has been determined in a Sr_2FeMoO_6-based tunnel junction device [5] and in $(BaSr)_2FeMoO_6$ granular samples [3]. Such a value can be attributed to *mis-site or antisite disorder* in which some Fe and Mo interchange their positions and is consistent both with a lower T_C and a saturation magnetization lower than the expected value of $4\mu_B$ in the ordered system. All samples present different degrees of disorder. The effect of disorder on the magnetization has been investigated in off-stoechiometric $Sr_2Fe_{1+x}Mo_{1-x}O_6$ $(-1 < x < 0.25)$ showing the depolarization of the conduction band [6]. The TMR is strongly reduced by the disorder and suppressed in highly disordered samples [7,8]. It can be inferred that disorder is detrimental to the half-metallic character, giving rise to up-spin states at the Fermi energy thus depolarizing the conduction band. In this letter, we aim to clarify the effect of disorder on the evolution of the density of states and the resulting spin polarization. We will show that the disorder-induced depolarization of the conduction band occurs through *new bonding states,* essentially with Mo character, appearing at the Fermi energy. This will emphasize that an electron-electron interaction on the Mo site, even small ~1 eV [9,10], is the key in determining a reasonable valence on Mo antisites and consequently, an appropriate electronic scheme with disorder.

The perfectly ordered lattice of Sr_2FeMoO_6 consists of alternating Fe and Mo along the three cubic axis of the perovskite structure. The nature of disorder may be quite complex, with the presence of different local environments and/or antiphase domains [11] for example, and may vary from sample to sample. Recent X-ray absorption fine structure (XAFS) experiments have revealed the persistence of short-range order even in samples with high degree of long-range disorder, suggesting the presence of nanosized antiphase domains [12]. In view of the complexity of such a nanoscopically inhomogeneous situation we adopt the simplest view of disorder defining the order parameter a $(1/2 < a < 1)$ as the probability to find Fe and Mo in their correct positions (S), respectively sublattices α and β of the ordered structure, *i.e.* $p^{\alpha}_{Fe} = p^{\beta}_{Mo} = a$, while the proportion of Fe and Mo in the wrong position or antisites (AS) are $p^{\beta}_{Fe} = p^{\alpha}_{Mo} = 1 - a$. In the presence of AS disorder, both Fe(AS)-O-Fe(S) and Mo(AS)-O-Mo(S) nearest-neighbors (n.n.) bonds are generated. Firstly, it has become clear that n.n. Fe-Fe local spins are antiferromagnetically coupled [6,13], so we consider all Fe local spins to be $\Uparrow$ (+) on sublattice α and $\Downarrow$ (−) for AS on sublattice β. On the other hand, n.n. Mo-Mo hopping channels are opened which are available for both up and down spins since electronic correlations on Mo sites are weak, thereby depolarizing the conduction band.

To account for disorder in these compounds, it is essential to consider the effect of the local environment on site energies. It has been pointed out [14] that Fe (Mo) AS electronic energies E'_{Fe} and E'_{Mo} should be different from site energies due to different environments. Basically, an electron on an AS-Fe will be surrounded by Fe^{3+} ions instead of Mo^{6+}, therefore experiencing a less attractive potential and driving the AS Fe valence towards 3+. This view is supported by the estimate of the diagonal energies from *ab initio* calculations [15], AS Fe is found to have an energy that is about 1 eV higher than the energy of the Fe at the regular site, but the effect of the environment seems however much weaker for Mo [16], so we will take $E'_{Mo} = E_{Mo}$. Considering a Madelung-like contribution from the nearest charges we can define and average AS Fe and write a linear dependence on a, $E'_{Fe} = E_{Fe} + \delta(2a - 1)$, $E_{Mo} = E_{Fe} + \Delta_0 - \delta(1 - a)$, Δ_0 represents the bare charge transfer energy in the ordered state; we shall take $\delta = w$ since $w = 1$ eV is a good estimate from band structure calculations.

In order to obtain the density of states, we follow the model of ref. [17] to calculate the local Green's functions. In the fully ordered case, half-metallicity has been understood on the basis of a strongly correlated description for Fe with the stable configuration $d^5(Fe^{3+})$ forming high-spin $S = 5/2$ localized spins $\vec{S}_i$ according to Hund's coupling, together with Mo^{6+} cores and conduction electrons (one per Mo) [17]. This electronic picture corresponds to the limit of large exchange integral J in the dynamical mean-field (DMF) approach of Chattopadhyay and Millis [18] and is consistent with the *ab initio* calculations [1,9,10], photoemission spectroscopes [19], X-ray magnetic circular dichroism experiment [20] for the ferromagnetic ground state of Sr_2FeMoO_6. Itinerant electrons coming from Mo can move around between Fe and Mo ions in the exchange split t_{2g} orbitals leading to an intermediate valence configuration for Fe and Mo. In ref. [17] it has been further considered that the local spins can be either parallel or antiparallel to the spin quantization axis of itinerant electrons. This simplified picture gives results for the Curie temperature which compare very well with those of DMF, performing an average over the orientation of the classical local spins [18]. The conduction band arising from the three degenerate t_{2g} orbitals is described by a tight-binding model. On Fe sites all the five d-orbitals of one spin channel being occupied, *the "double-exchange interaction" simply reduces to constraints on the Fe-Mo hopping*: itinerant electrons can hop to Fe sites only with an antiparallel orientation with respect to the localized spins. On the other hand, Mo is not a strongly correlated metal and usually non magnetic, the interatomic correlation and exchange within d states being small compared to the $3d$ transition metals [21], therefore there is no constraint for electrons hopping on Mo sites. The Fe-Mo hopping stabilizes a ferromagnetic arrangement of the local spins and also leads to the opposite spin polarization of the conduction electrons in the fully ordered Sr_2FeMoO_6 [17]. The robustness of the ferromagnetic state is determined by the Fe-Mo charge transfer energy and the Fe-Mo hopping t [9,17,22]. In an alternating Bethe lattice in the limit of infinite coordination z and zt^2 scaled as $w^2/4$, $z=4$ (see footnote [1]) and w defining half the corresponding bandwidth, the local average Green's functions take the DMF form $G^{-1}_{ii,\sigma}=\omega-\varepsilon_i-\sum_{l\neq i}t^2G_{ll,\sigma}$, ε_i being the corresponding site energies and the summation is over all n.n. sites. In the presence of disorder this requires to take an average over atomic configurations. In general, the disordered state has both diagonal (different sites energies) and off-diagonal (different hopping parameters) disorder. Our averaging procedure over n.n. is similar to the one adopted by Sarma *et al.* [23] for diagonal disorder only and is equivalent to the coherent potential approximation using the locator formalism [24]. This represents the simplest description of a nanoscopic inhomogeneous situation. However, short-range order could be included requiring a precise knowledge of its nature, but we believe that this will not modify qualitatively our results. The σ electrons local average Green's functions $G^{\gamma Fe}_{\sigma\mu}$ and $G^{\gamma Mo}_{\sigma}$ for Fe and Mo on $\gamma=\alpha,\beta$ sublattice are given by

$$G^{\alpha Fe}_{\downarrow +}=\frac{1}{\omega-\widetilde{E}_{Fe}-\frac{w^2}{4}aG^{\beta Mo}_{\downarrow}},\tag{1}$$

$$G^{\beta Mo}_{\downarrow}=\frac{1}{\omega-\widetilde{E}_{Mo,\downarrow}-\frac{w^2}{4}aG^{\alpha Fe}_{\downarrow +}-\frac{w'^2}{4}(1-a)G^{\alpha Mo}_{\downarrow}},\tag{2}$$

$$G^{\alpha Mo}_{\downarrow}=\frac{1}{\omega-\widetilde{E}'_{Mo,\downarrow}-\frac{w'^2}{4}aG^{\beta Mo}_{\downarrow}}\tag{3}$$

and

$$G^{\beta Fe}_{\uparrow -}=\frac{1}{\omega-\widetilde{E}'_{Fe}-\frac{w^2}{4}(1-a)G^{\alpha Mo}_{\uparrow}},\tag{4}$$

$$G^{\alpha Mo}_{\uparrow}=\frac{1}{\omega-\widetilde{E}'_{Mo,\uparrow}-\frac{w^2}{4}(1-a)G^{\beta Fe}_{\uparrow -}-\frac{w'^2}{4}aG^{\beta Mo}_{\uparrow}},\tag{5}$$

$$G^{\beta Mo}_{\uparrow}=\frac{1}{\omega-\widetilde{E}_{Mo,\uparrow}-\frac{w'^2}{4}(1-a)G^{\alpha Mo}_{\uparrow}},\tag{6}$$

$\mu=\pm$ reminds the Fe local spins orientation on each sublattice. Here, the Green's functions refer to a single orbital. We consider only the nearest-neighbor hoppings between the two different sublattices. w and w' being the bandwidths corresponding to Fe-Mo and Mo-Mo hopping respectively, $w'=qw>w$ due to the larger extension of the Mo 5d states; $q\sim 2$–2.5 appears reasonable and compatible with some *ab initio* calculations, we shall take $q=2$. We neglect Fe-Fe hopping which is expected to be much smaller. The number of carriers per unit cell is $n=1$. $\widetilde{E}_{Fe}$, $\widetilde{E}_{Mo,\sigma}$ are effective Fe and Mo site energies on the right sublattice taking into account electronic correlations, while $\widetilde{E}'_{Fe}$ and $\widetilde{E}'_{Mo,\sigma}$ are the energies when they occupy AS positions. As we mentioned, it is fundamental to take into account correlations U_{Mo} on Mo, so, to be consistent, we need to include also the stronger correlations among itinerant electrons on Fe sites adding H^{Mo}_c and H^{Fe}_c:

$$H^{Mo}_c=(U^{Mo}+2J^{Mo})\sum_{i,\nu}n_{i\nu\uparrow}n_{i\nu\downarrow}+U^{Mo}\sum_{i,\nu,\nu'\neq\nu}n_{i\nu\uparrow}n_{i\nu'\downarrow}$$
$$+(U^{Mo}-J^{Mo})\sum_{i,\nu,\nu'\neq\nu,\sigma}n_{i\nu\sigma}n_{i\nu'\sigma},\tag{7}$$

$$H^{Fe}_c=(U^{Fe}-J^{Fe})\sum_{j,\nu,\nu'\neq\nu,\sigma}n_{j\nu\sigma}n_{j\nu'\sigma},\tag{8}$$

i,j corresponding to sites occupied by Mo and Fe irrespective of the sublattice, ν and ν' label the three t_{2g} orbitals. On Fe sites, only one spin direction being possible intervene only $U^{Fe}_{eff}=(U^{Fe}-J^{Fe})\sim 2$–4 eV. Contrary to $\widetilde{E}_{Mo,\sigma}$ and $\widetilde{E}'_{Mo,\sigma}$, $\widetilde{E}_{Fe}$ and $\widetilde{E}'_{Fe}$ do not depend on the spin since only one spin direction is possible on each Fe site. Site energies are calculated self-consistently using mean-field approximation to treat the correlations *i.e.*

$$\widetilde{E}_{Fe}=E_{Fe}+\frac{2}{3}U^{Fe}_{eff}\left\langle n^{Fe}_{\alpha\downarrow}\right\rangle,\tag{9}$$

$$\widetilde{E}'_{Fe}=E'_{Fe}+\frac{2}{3}U^{Fe}_{eff}\left\langle n^{Fe}_{\beta\uparrow}\right\rangle,\tag{10}$$

$$\widetilde{E}_{Mo,\sigma}\;=\;E_{Mo}+(U^{Mo}+\frac{2}{3}J^{Mo})\left\langle n^{Mo}_{\beta,-\sigma}\right\rangle$$
$$+\frac{2}{3}U^{Mo}_{eff}\left\langle n^{Mo}_{\beta,\sigma}\right\rangle\tag{11}$$

and

$$\widetilde{E}'_{Mo,\sigma}\;=\;E'_{Mo}+(U^{Mo}+\frac{2}{3}J^{Mo})\left\langle n^{Mo}_{\alpha,-\sigma}\right\rangle$$
$$+\frac{2}{3}U^{Mo}_{eff}\left\langle n^{Mo}_{\alpha,\sigma}\right\rangle,\tag{12}$$

where we used the orbital degeneracy $\langle n_{i\nu\sigma}\rangle=\frac{\langle n_{i\sigma}\rangle}{3}$ and $U^{Mo}_{eff}=U^{Mo}-J^{Mo}$. We shall take $U^{Fe}_{eff}=3w$, $J^{Mo}=0.1w$ and we can vary U^{Mo}_{eff}. Evidently, the effective charge transfer energy determining the density of states and electronic properties depends self-consistently on the correlations and disorder. It is convenient to take the charge transfer in the ordered state ($a=1$) as a reference (with superscript 0 for the corresponding quantities)

$$\Delta\;=\;\widetilde{E}^0_{Mo,\downarrow}-\widetilde{E}^0_{Fe}=$$
$$E_{Mo}-E_{Fe}+\frac{2}{3}U^{Mo}_{eff}\left\langle n^{Mo}_{\beta,\downarrow}\right\rangle^0-\frac{2}{3}U^{Fe}_{eff}\left\langle n^{Fe}_{\alpha\downarrow}\right\rangle^0.\tag{13}$$

It has been shown that small charge transfer is realized in Sr_2FeMoO_6 [17], so we consider here $\Delta=0$ to reproduce the mixed-valence character $Fe^{2.5+}$ and $Mo^{5.5+}$ as evidenced experimentally [25,26].

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[1] In the cubic symmetry, due to the symmetry of the intermediate O orbitals, the first and second neighbor hoppings are nonzero only between orbitals of the same t_{2g} symmetry $\gamma=xy$, yz, zx and only between orbitalslying in the corresponding plane γ; it is the same for the three orbitals, thus giving three degenerate two-dimensional bands with coordination $z=4$.

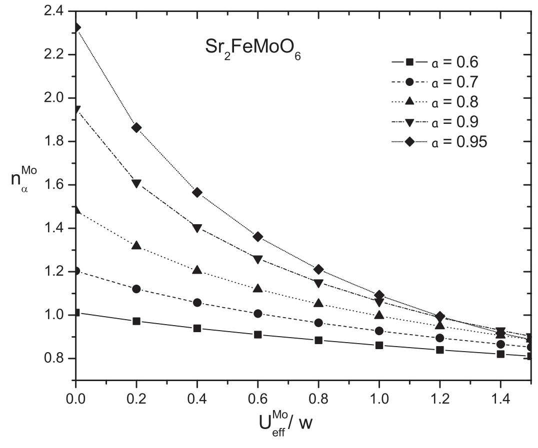

Fig. 1: The total electron occupation on Mo antisites n_α^{Mo} as a function of Mo correlation U_{eff}^{Mo}/w for different values of the disorder. $n=1$ and the charge transfer energy $\Delta=0$.

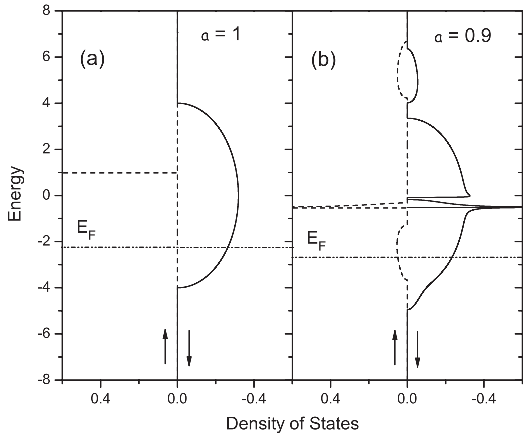

Fig. 2: Evolution of the density of states with disorder for $U_{eff}^{Mo}=w$ (other parameters are the same as in fig. 1) (a) represents the half-metallic fully polarized ordered case and (b) illustratres a disordered situation ($a=0.9$) showing the appearence of Mo(S)-Mo(AS) bonding states at E_F for the two spin channels. We take $\widetilde{E}_{Fe}^0=0$ as the energy reference.

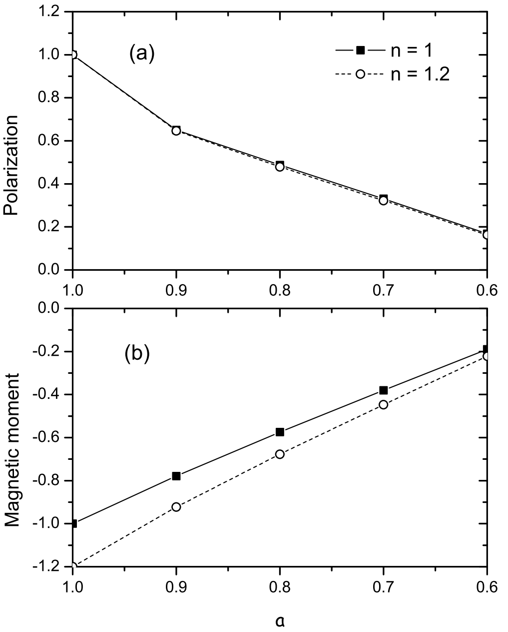

Fig. 3: The spin polarization P (a) and the itinerant magnetic moment μ (b) as function of the disorder for different band fillings n (electron doping). (Others parameters are the same as in fig. 2.)

Our calculated density of states describes correctly the ordered half-metallic fully polarized state with the partially filled Fe-Mo down-spin minority band (fig. 2(a)). The unoccupied up-spin level $\widetilde{E}_{Mo,\uparrow}$ corresponds to the narrow band seen in the band structure calculations, the finite bandwidth being due to the next n.n. Mo-Mo hopping within sublattice β not included in our calculations. The weak disorder limit ($a\to 1$) points towards the importance of Mo correlations. From the ↑ channel Mo Green's functions (5), (6) we obtain Mo(S)-Mo(AS) bonding and antibonding localized levels with weight $(1-a)$. Neglecting correlations [27] the bonding level lies below the Fermi level and will be occupied, providing 3/2 electrons per AS-Mo; this occurs similarly for ↑ electrons but Mo states are coupled with Fe states and the Mo occupation is slightly less, nevertheless the total AS-Mo occupation is 2.73 giving an unlikely valence 3.27+ for these AS Mo. With Mo correlations the bonding states are pushed to higher energies reducing the electron occupation on Mo(AS) as shown in fig. 1; we see that typical values of $U_{eff}^{Mo}\sim$ 1–1.2 eV [9] provides a very good agreement for $a\simeq 0.9$ with the experimental estimate of Mo(AS) valence $\sim 5+$ [28]. So we adopt $U_{eff}^{Mo}=w$ in the following to present a consistent electronic picture with disorder. The evolution of the density of states with the amount of disorder is shown in fig. 2 (energies are in units of t, $w=4$). Figure 2(a) represents the density of states for the ordered half-metallic state ($a=1$). Figure 2(b) illustrates the behavior with disorder $a=0.9$. The bonding and antibonding states present in the dilute disorder limit transforms into bands. These *new* small bands are particularly well defined in the spin-↑ channel, they are only weakly mixed with the Fe(AS) states at $\widetilde{E}'_{Fe}$ and have a spectral weight $(1-a)$. These states are also present in the spin-↓ channel at the bottom and the top of the main Fe-Mo band but there is a stronger admixture with Fe(S)-Mo(S) states. *The spin depolarization of the conduction band occurs essentially through these partially filled new Mo(S)-Mo(AS) bonding states*, a scenario in contrast with the rigid band shift model recently proposed by Rubi *et al.* [29]. We also note that the Fe(AS) states above the Fermi level are weakly admixed with Mo states so the valence of Fe antisites remains $\sim 3+$ in full agreement with the value assigned to fit Mössbauer spectra [25].

We calculate the two key quantities: the spin polarization P and the itinerant magnetic moment $\mu=n_\downarrow-n_\uparrow$ shown respectively in fig. 3(a) and fig. 3(b). As expected both decrease with increasing disorder, however P is more sensible to a small disorder than μ; μ presents a quasi-linear decrease. Again good agreement with experimental results is obtained. For example, for samples with estimated order $a \sim 0.9$–0.92 we obtain $P \sim 0.65$–0.7 which compares well the 0.6 value reported by Rubi *et al.* [29], and $\mu \sim 0.8$ comparable with the value given by Topwal *et al.* [6]. It is worth mentioning a few words on electron-doped systems $A_{2-x}L_xFeMoO_6$ (A is divalent Sr or Ca and L is a trivalent lanthanide) in which $n = 1 + x$. The polarization decreases upon trivalent substitution [29]. Our calculations indicate that the polarization does not seem to depend much on n itself at least for small doping as fig. 3 shows for $n = 1.2$. This leads us to infer that the decrease of the polarization upon electron doping reflects the increase of disorder rather than the number of carriers.

In conclusion, the present results illustrate how missites disorder affects the electronic density of states by introducing new Mo(S)-Mo(AS) bonding states at the Fermi level for the two spin channels. As a consequence, the system loses its half-metallic character and the spin polarization as well as the magnetic moment of itinerant electrons are inevitably reduced. We have shown the key role played by Mo interatomic correlations, even moderate, in determining the valence of Mo antisites. An overall good agreement is obtained for Fe and Mo valences in regular and antisite positions. Although it has been specifically applied to the Fe-Mo double-perovskite family, our picture has a more general validity for other systems with different charge transfer energies.

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