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Ab initio study of disorder effects on the electronic and magnetic structure of Sr_2FeMoO_6

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We have investigated the electronic structure of ordered and disordered Sr_2FeMoO_6 using *ab initio* band-structure methods. The effect of disorder was simulated within supercell calculations to realize several configurations with mis-site disorders. It is found that such disorder effects destroy the half-metallic ferromagnetic state of the ordered compound. It also leads to a substantial reduction of the magnetic moments at the Fe sites in the disordered configurations. Most interestingly, it is found for the disordered configurations that the magnetic coupling within the Fe sublattice as well as that within the Mo sublattice always remain ferromagnetic, while the two sublattices couple antiferromagnetically, in close analogy to the magnetic structure of the ordered compound, but in contrast to recent suggestions.

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I. INTRODUCTION

Discovery of a large negative magnetoresistance, namely, the colossal magnetoresistance[1] (CMR) in doped manganites has attracted a great deal of attention in recent years, because of the technological importance of these materials being used as magnetic devices.[2] The pronounced influence of the low magnetic field on the resistance of these compounds is believed to be caused by the high degree of spin polarization of the charge carriers, arising due to the half-metallic ferromagnetic nature of these materials below the magnetic transition temperature T_c. However the transition temperatures obtained in the case of the manganites were low for room-temperature applications, leading to a further search for half-metallic oxides with much higher T_c. Recently Kobayashi *et al.*[3] reported a large magnetoresistance effect with a fairly high magnetic transition temperature of about 418 K in Sr_2FeMoO_6, a material belonging to the class of double perovskites ($A_2BB'O_6$), where the alkaline-earth ion A is Sr and transition-metal ions B and B' are Fe and Mo arranged in the rock-salt structure. Such a high magnetic ordering temperature, which is higher than that in the manganites (250 K–350 K), is surprising and has been recently explained in terms of a new mechanism based on a novel magnetic interaction.[4]

In the idealized ordered structure of these double perovskite systems, the two transition-metal ions B and B' are arranged alternately along the cubic axes in all three directions. Specifically, in the case of Sr_2FeMoO_6 each of the two transition-metal sites, namely, the Fe^{3+} ($3d^5$, $S=\frac{5}{2}$) and Mo^{5+} ($4d^1$, $S=\frac{1}{2}$) sites, are believed to be ferromagnetically arranged within each sublattice, while the two sublattices are supposed to be coupled antiferromagnetically, giving rise to a $S=2$ state. Recently, it has been found that it is possible to synthesize samples of Sr_2FeMoO_6 with substantial mis-site disorders where Fe and Mo sites interchange their positions.[5,6] Such a disorder is found to have profound effects on the physical properties of this compound, particularly in terms of its magnetic and CMR properties. It appears that such a disorder may even play a role in determining the properties of the so-called *ordered* system,[7] since a finite amount of disorder is found to be present also in such samples. Rietveld analysis of the x-ray powder-diffraction data of Kobayashi *et al.* indicated 87% order of Fe and Mo ions. The saturation moment observed by Kobayashi *et al.*[3] at 4.2 K was lower than the expected value of $4\mu_B$ per formula unit (f.u.) considering the antiferromagnetic coupling between Fe^{3+} and Mo^{5+} ions, which is indicative of, as pointed out by the authors, the mis-site-type disorder of the B-site rock-salt arrangement. Later, the study by Sarma *et al.*,[5] which reported ordering at the Fe and Mo sites in their sample to be 91%, slightly higher than that of Kobayashi *et al.*, showed a sharper low-field response with improved magnetoresistive behavior. This may be related to the effect of disorder on the half-metallic state of the compound, which is considered to be central to the low-field magnetic behavior via the spin-dependent scattering processes, as mentioned earlier. The above studies are for bulk-sintered samples. Further studies on thin films of Sr_2FeMoO_6 show the saturation magnetization and the magnetoresistive behavior to be very different for the films grown under different conditions.[8,9] The magnetization is reduced substantially for samples having random arrangements of Fe, Mo sublattices. The reduction in the net magnetization as measured in the experiment can occur by two distinctly different routes. One possibility is that the disorder destroys the specific spin arrangement of the Fe and Mo sites without any significant effect on the individual magnetic moments at these sites. For example, disorder may lead to antiferromagnetic couplings between different Fe sites,[10] instead of the ferromagnetic coupling proposed for the idealized ordered structure. Alternately, the magnetic moments at each individual site may decrease due to the different chemical environment induced by the disorder, without any effect on the nature of the spin ordering. The real situation may even be a combination of both these effects, with a simultaneous reduction in the magnetic moments at different sites as well as a change in the nature of the magnetic coupling between different sites. De-

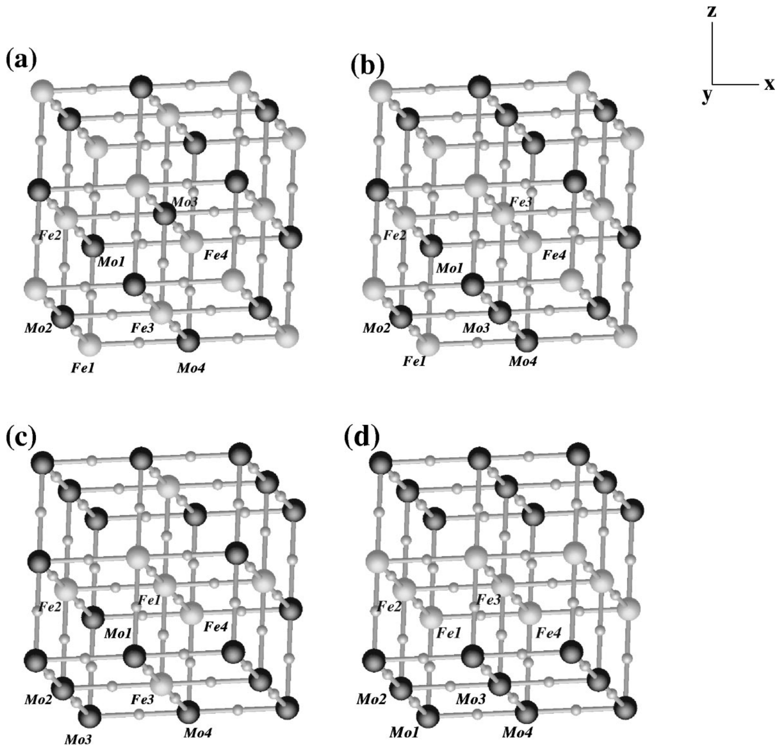

FIG. 1. Structure of ordered and disordered Sr_2FeMoO_6. Disordered structure is shown for three different model configurations. In the first two configurations [(b) and (c)] one pair of Mo and Fe have been interchanged, while in the third configuration [(d)] two pairs have been interchanged. Large gray and black circles indicate Fe and Mo sites while the small gray circles represent sites occupied by O atoms.

tailed knowledge about the disordered compound is therefore important for better understanding of the various experimental results and designing of samples with better magnetic and magnetoresistive properties. This motivated the present study, where we have employed the state of the art first-principles band-structure method to study the electronic structure and hence the magnetic properties of the ordered and disordered Sr_2FeMoO_6. Our results clearly support the view that individual magnetic moments at each Fe site are strongly reduced due to the destruction of the half-metallic state in the presence of disorder, while the coupling between the various Fe sites continues to be ferromagnetic, in contrast to a recent suggestion.[10]

II. METHODOLOGY

Sr_2FeMoO_6 occurs in the body-centered tetragonal structure with a space group of $I4/mmm$ and lattice constants $a = b = 5.57$ Å and $c = 7.90$ Å.[3] The oxygen atoms surrounding the Fe and Mo sites provide the octahedral environment. The FeO_6 and MoO_6 octahedra alternate along the three cubic axes, while Sr atoms occupy the hollow formed by the corners of FeO_6 and MoO_6 octahedra at the body-centered positions. In Fig. 1(a) we show the four-formula-unit supercell of the structure. To simplify the view only Fe, Mo, and O atoms are shown. The alternate positioning of transition-metal Fe and Mo sites is evident from this figure.

We have employed the linear muffin-tin orbital (LMTO) method, implemented in the basis of screened and energy-independent muffin-tin orbitals to study the electronic structure of the ordered and disordered Sr_2FeMoO_6 compounds. The effect of disorder has been modeled by supercell calculations. The calculations have been performed within the framework of atomic sphere approximation (ASA), so that the muffin-tin spheres were replaced by space-filling atomic spheres. A detailed description of the LMTO-ASA formalism can be found elsewhere.[11] The partitioning of the space into atom-centered spheres was carried out ensuring that the overlaps between different atomic spheres do not exceed 16%. The basis set consisted of *s*, *p*, *d*, and *f* on Sr and Mo sites and *s*, *p*, and *d* on Fe and O sites. The *s* and *p* channels on the Sr site; *d* and *f* channels on the Mo site; *s*, *p*, and *d* channels on the Fe site; and *p* channels on the O site were considered to be active channels, while the rest were downfolded. The scalar relativistic spin-polarized calculations with (6 6 6) **k** points were carried out for the self-consistency. The exchange-correlation part of the potential was treated using the pure gradient expansion of the generalized gradient approximation (GGA). The use of the GGA was found to be crucial to obtain the correct magnetic ground state of the compound, indicating the possible importance of correlation effects in this compound. We have calculated the electronic structures of both ordered and disordered systems in order to have a complete comparative study within the spin-polarized approach.

In order to compare the electronic structures of the ordered and the disordered Sr_2FeMoO_6 on the same footing, calculations were performed by constructing supercells of four formula units with four inequivalent Fe sites at (0, 0, 0), (0, 0.5, 0.5), (0.5, 0.5, 0), and (0.5, 0, 0.5) positions and four inequivalent Mo sites at (0, 0, 0.5), (0, 0.5, 0.0), (0.5, 0.5, 0.5), and (0.5, 0, 0) positions as shown in Fig. 1(a). These inequivalent Fe and Mo sites are designated as Fe1, Fe2, Fe3, and Fe4 and Mo1, Mo2, Mo3, and Mo4, respectively. While these four sites are crystallographically equivalent for the ordered sample, they are symmetry distinct in the presence of mis-site disorders. The effect of positional disor-

TABLE I. Coordination of Mo and Fe sites surrounding each inequivalent Fe and Mo site for different neighbor shells for the ordered and three disordered configurations. The shells are marked with distances and the vectors connecting the sites. Due to the slight deviation from the cubic symmetry the vectors involving translations along the *c* axis have slightly different distances than those involving translations along the *a* and *b* axes. The last two columns give the magnetic moment at various inequivalent sites and the net magnetic moment.

	Sites	Distance = 3.94 Å $[\frac{1}{2}\{010\}]$	Distance = 3.95 Å $[\frac{1}{2}\{001\}]$	Distance = 5.57 Å $[\frac{1}{2}\{110\}]$	Distance = 5.58 Å $[\frac{1}{2}\{101\}]$	Magnetic moment (μ_B)	Net moment per f.u. (μ_B)
Ordered	Fe	4 Mo	2 Mo	4 Fe	8 Fe	3.8	4.0
	Mo	4 Fe	2 Fe	4 Mo	8 Mo	−0.3	
Disordered (configuration 1)	Fe 1	4 Mo	2 Mo	4 Mo	8 Fe	3.8	3.1
	Fe 2	2 Fe+2 Mo	2 Mo	4 Fe	4 Fe+4 Mo	3.1	
	Fe 3	4 Fe	2 Mo	4 Mo	8 Mo	2.0	
	Fe 4	2 Fe+2 Mo	2 Mo	4 Fe	4 Fe+4 Mo	3.1	
	Mo 1	4 Fe	2 Fe	4 Fe	8 Mo	−0.3	
	Mo 2	2 Mo+2 Fe	2 Fe	4 Mo	4 Mo+4 Fe	−0.3	
	Mo 3	4 Mo	2 Fe	4 Fe	8 Fe	−0.2	
	Mo 4	2 Mo+2 Fe	2 Fe	4 Mo	4 Mo+4 Fe	−0.3	
Disordered (configuration 2)	Fe 1	4 Fe	2 Fe	4 Mo	8 Mo	2.5	3.2
	Fe 2	2 Fe+2 Mo	2 Mo	4 Fe	4 Fe+4 Mo	3.1	
	Fe 3	4 Mo	2 Fe	4 Mo	8 Fe	3.1	
	Fe 4	2 Fe+2 Mo	2 Mo	4 Fe	4 Fe+4 Mo	3.1	
	Mo 1	4 Fe	2 Mo	4 Fe	8 Mo	−0.3	
	Mo 2	2 Fe+2 Mo	2 Fe	4 Mo	4 Mo+4 Fe	−0.3	
	Mo 3	4 Mo	2 Mo	4 Fe	8 Fe	−0.1	
	Mo 4	2 Mo+2 Fe	2 Fe	4 Mo	4 Mo+4 Fe	−0.3	
Disordered (configuration 3)	Fe	4 Fe	2 Mo	4 Fe	8 Mo	2.5	2.5
	Mo	4 Mo	2 Fe	4 Mo	8 Fe	−0.3	

der at Fe/Mo sites is simulated by interchanging the Fe and Mo sites, so as to generate different chemical environments surrounding each inequivalent Fe and Mo site while keeping the concentration of Fe/Mo sites fixed. We have performed the disordered calculation for three different disordered configurations, so as to check that the result is not influenced by the specific choice of a particular disordered configuration. The first configuration is obtained by exchanging the Fe3 and Mo3 sites at (0.5, 0.5, 0) and (0.5, 0.5, 0.5) positions, respectively, that is, by interchanging one of the Fe-Mo pairs that is the nearest neighbor to each other. The second configuration is obtained by interchanging Fe1 and Mo3 at (0, 0, 0) and (0.5, 0.5, 0.5), respectively; this amounts to an interchange of the distant Fe-Mo pairs. Figures 1(b) and 1(c) show the corresponding structures with an interchange of only one Fe-Mo pair. The structure shown in Fig. 1(b) can be derived from the structure shown in Fig. 1(a) by replacing the Mo site at the body center with an Fe atom and the face-centered Fe sites in *ab* planes with Mo. Similarly the structure in Fig. 1(c) is derived by replacing the Mo site at the body center with Fe and the Fe sites at the corners with Mo. Figure 1(d) shows the configuration that can be derived from the configuration shown in Fig. 1(b) by an additional exchange between Fe1 and Mo1. These replacements in turn lead to the distribution of different environments in terms of Fe and Mo sites for different inequivalent Fe and Mo sites.[12] Table I summarizes the distribution of Fe and Mo neighbors at various shells for different inequivalent Fe and Mo sites in each of these structures.

III. RESULTS AND DISCUSSION

The spin-polarized LMTO-GGA calculation for the ordered structure gives the ground state to be half-metallic ferrimagnetic with a finite density of states in the spin-down channel and a zero density of states in the spin-up channel and antiferromagnetic ordering between the high-spin $3d^5$ ($S=\frac{5}{2}$) Fe^{3+} ion and the $4d^1$ ($S=\frac{1}{2}$) Mo^{5+} ion, in agreement with previous results.[3,4] The magnetic moment at the Fe site was found to be $+3.8\mu_B$ and that at the Mo site $-0.3\mu_B$ in good agreement with the pseudopotential calculation.[3] The six oxygen sites altogether contribute a magnetic moment of about $0.5\mu_B$, thereby making the total magnetic moment of the system to be $4\mu_B$ per f.u. The magnetic moment at the oxygen sites accounts for the missing moments at the Fe and Mo sites considering their Fe^{3+} and Mo^{5+} configurations, which in turn indicates a strong covalency effect present in the system.

In Fig. 2(a) we show the calculated total density of states (DOS) and various partial densities of states for $\mathrm{Sr_2FeMoO_6}$. The corresponding band dispersions along the high-symmetry lines are shown in Fig. 2(b). It is to be noted that

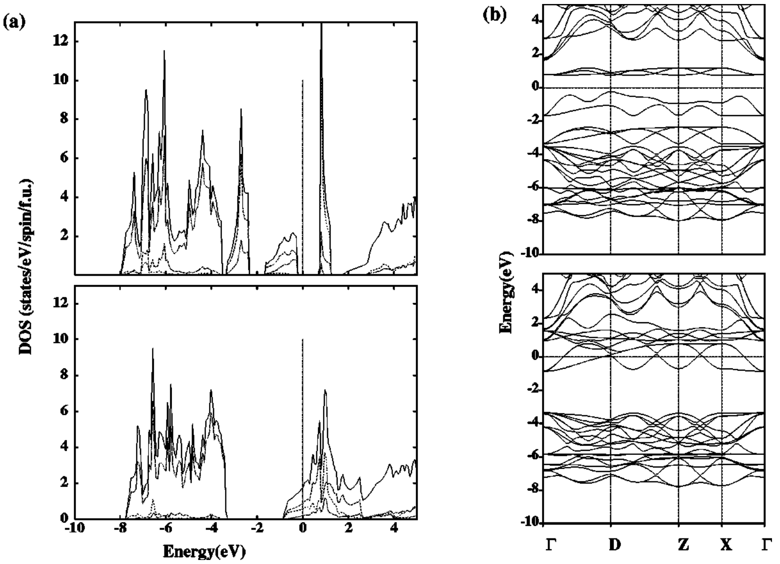

FIG. 2. (a) Total and partial densities of states of ordered Sr_2FeMoO_6. The solid line represents the total density of states, while long-dashed, dashed, and dotted lines represent Fe-*d*, Mo-*d*, and O-*p* partial densities of states, respectively. (b) Band dispersion of ordered Sr_2FeMoO_6 along the high-symmetry lines. The zero of the energy marked by the dashed line has been set at the Fermi energy.

the band dispersions are plotted along the symmetry directions of the original primitive unit cell and not of the supercell. The top panels are for the spin-up channel and the bottom panels are for the spin-down channel. Since the system orders ferrimagnetically with antiferromagnetic coupling between Fe^{3+} and Mo^{5+} ions, the spin-up channel is the majority channel for Fe and the minority for Mo and vice versa for the spin-down channel. The predominant feature in the band dispersion is a band gap of about 0.8 eV in the spin-up channel while bands cross the Fermi level (E_F) in the spin-down channel, resulting in the half-metallic state.

For our discussion of the density of states, which is limited to an energy window of -8 eV to about 2 eV, we shall be primarily concerned with the Fe-*d*, Mo-*d*, and O-*p* states, since the Sr-derived states appear higher in energy. As is evident from the partial density of states due to oxygen in the spin-up and spin-down channels, the states spanning -8 eV to -3.5 eV, separated from the rest of the states by a gap of about 0.2 eV in the spin-up channel and about 2.5 eV in the spin-down channel, are primarily contributed by oxygen, while it has some contribution to states appearing close to the Fermi energy, mixed with Fe-*d* and Mo-*d* states. The presence of an approximate cubic symmetry of the octahedral coordination of the oxygen atoms around the transition-metal Fe and Mo sites results in a splitting of the *d* levels of Fe and Mo sites into triply degenerate t_{2g} orbitals with lower energy and doubly degenerate e_g orbitals with higher energy. The Fe-*d* DOS in the spin-up channel span the energy from -3.3 eV to -2.3 eV and from -1.7 eV to -0.2 eV, contributed by the crystal-field-split t_{2g} and e_g states, respectively, giving the crystal-field splitting of ≈ 2 eV. With the Fe ion being in the high-spin d^5 state, the spin-up Fe states are completely filled, lying below the Fermi energy. In the spin-down channel the splitting between t_{2g} and e_g states disappears due to a large bandwidth of Fe t_{2g} states, which primarily comes from the hybridization with Mo t_{2g} states[4] via oxygen states. The t_{2g} and e_g resolved partial density of states (not shown here) shows the exchange splitting between the spin-up and spin-down Fe t_{2g} and e_g contributions to be about 3.5 eV. The Mo-*d* density of states in the spin-up channel between an energy range of 0.7 eV to 1.2 eV is predominantly of the t_{2g} type, which is unusually narrow due to the large energy difference between the Fe and Mo t_{2g} levels in the spin-up channel, substantially suppressing the hopping processes. The crystal-field-split spin-up e_g levels appear higher in energy, starting from 3 eV. Down-spin Mo t_{2g}, due to the hybridization with oxygen *p* and Fe t_{2g} in particular, as mentioned before, acquires a large bandwidth spanning an energy range of 3 eV, from -1 eV to 2 eV, thereby crossing the Fermi level, while the empty e_g states, as in the case of the spin-up channel, are higher in energy. The exchange splitting between the up and down Mo t_{2g} and e_g states is much smaller (≈ 0.5 eV) and in the opposite direction as that of Fe, in agreement with the magnetic-moment results. It should be noted here that though the exchange splitting of the Mo $4d$ band is much smaller than that (≈ 3.5 eV) of the Fe $3d$ band, these results surprisingly suggest comparable Hund's coupling strengths in the Fe $3d$ and Mo $4d$ manifolds, as suggested earlier.[4] Since the exchange splitting of the band is given by the product of the coupling strength and the number of unpaired electrons in that band, the coupling strength in Fe $3d$ is in the order of $3.5/5 = 0.7$ eV, whereas in the Mo $4d$, it is ≈ 0.5 eV.

Our self-consistent field calculations for the disordered models yield the ground state to be magnetic with a reduced calculated magnetic moment per formula unit ($3.3\mu_B$ for the first configuration, $3.2\mu_B$ for the second configuration, and $2.5\mu_B$ for the third configuration) compared to that of the fully ordered sample ($4\mu_B$), establishing a pronounced and systematic effect of disorder on the magnetic structure of this system; the calculated results are also in agreement with the experimental observation of a lower magnetic moment with decreasing order in such systems.[13] In order to understand the microscopic origin of this reduction in the magnetic moment, we discuss below our results in some detail.

The ground-state magnetic structures for all the studied disordered configurations were found to retain the *ferrimagnetic* state of the ordered sample in the following sense. The

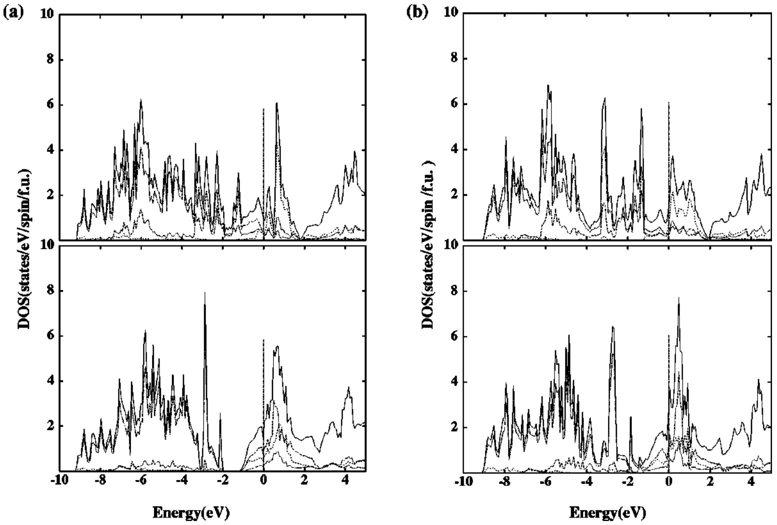

FIG. 3. Total and partial densities of states of disordered Sr_2FeMoO_6. Densities of states corresponding to the first and third configurations are shown in (a) and (b). As before, the solid line represents the total density of states, while long-dashed, dashed, and dotted lines represent Fe-*d*, Mo-*d*, and O-*p* partial densities of states, respectively. Top panels correspond to spin-up and bottom panels correspond to spin-down channels. The zero of the energy marked by the vertical dashed line has been set at the Fermi energy.

moments at each of the Fe sites were found to be ferromagnetically aligned, irrespective of the near-neighbor coordinations; the same is also found to be true for the Mo sites in every case (see Table I). The two sublattices of Fe and Mo are found to be antiferromagnetically coupled, giving rise to the ferrimagnetic state in every disordered configuration studied. This is in clear contrast to a previous Monte Carlo study[10] of a spin model, with local exchange interactions parametrized with antiferromagnetic coupling between the nearest-neighbor transition-metal sites, irrespective of the specific occupancies at these sites. Not surprisingly, this model suggests antiferromagnetically coupled Fe-O-Fe bonds, whenever two Fe ions form near-neighbor pairs. On the other hand, our results clearly show that the Fe-O-Fe interaction is actually *ferromagnetic*, i.e., for the first configuration, the Fe2, Fe3, and Fe4 sites, which are coordinated by Fe sites in the nearest-neighbor cation shell, as shown in Table I. This holds true even for the case of the Fe1 site in the second configuration, where all six nearest-neighboring cations along the *x*, *y*, and *z* directions are occupied by Fe. Thus, the experimentally observed reduction of the magnetic moment in disordered samples does not arise from an antiferromagnetic coupling of Fe-O-Fe bonds in a disordered sample; instead the reduction in the net magnetic moment is caused by the reduction of individual magnetic moments at different inequivalent Fe sites depending upon the various distributions of the number of Fe and Mo sites in the neighbor shells. Our full-fledged *ab initio* calculations therefore establish a very different behavior than that obtained from the previous model calculation, where the exchange couplings were *assumed* to be inherently antiferromagnetic for the nearest-neighbor Fe-O-Fe arrangements.

As Table I shows for the disordered configurations, the number of Fe and Mo atoms in various neighbor shells differ from that of the ordered arrangement. As expected the deviation is largest for Fe and Mo sites that are interchanged in position. We notice that the magnetic moments at the Fe sites are strongly influenced by the number of the Mo atoms in the nearest-neighbor shell. The higher the number of Mo neighbors surrounding each Fe atom, the larger the magnetic moment. When the Fe site is surrounded by only Mo sites in the nearest cation shell the magnetic moment at the Fe site in the disordered configuration is $3.8\mu_B$, the same as that of the ordered calculation. Reducing the number of Mo near neighbors by two from that of the ordered arrangement reduces the magnetic moment to $3.1\mu_B$, as can be seen for Fe2 and Fe4 in configuration 1 and Fe2, Fe3, and Fe4 in configuration 2 given in Table I. Reducing the number of Mo coordinating a central Fe even further decreases the magnetic moment at Fe sites down to $2.0–2.5\mu_B$. The reduction of the magnetic moment at the Fe sites with the reduction in Mo neighbors may be explained by considering the fact that the charge-transfer energy between the Fe and Mo is substantial. As a result the presence of Mo neighbors decreases the hopping probability from the Fe site, enhancing the importance of electron correlation and stabilizing a larger magnetic moment. On the contrary, the reduction in the number of Mo neighbors and the increase in the number of Fe neighbors has an opposite effect; the hopping to neighboring Fe sites enhances the bandwidth, making the central Fe site less correlated with a reduced magnetic moment.

In Figs. 3(a) and 3(b), we show the DOS and the partial Fe-*d*, Mo-*d*, and O-*p* DOS for the disordered configurations 1 and 3; the results from configuration 2 are very similar to those from configuration 1. The most evident consequence of the mis-site disorder that is common in all the results is a destruction of the half-metallic ferrimagnetic state due to the collapse of the gap present between the Fe e_g and Mo t_{2g} spin-up states in the ordered sample. Since the disorder has been introduced between Fe and Mo sublattices, the high-lying Sr-derived states at energy 2 eV and above remain more or less unaffected. The energy states lying low in energy, from about -9 eV to -4 eV in the spin-up channel and from -9 eV to -2 eV in the spin-down channel, are dominated by oxygen-*p* states while Fe *d* and Mo *d* contribute to states close to E_F in close similarity with the gross features of the ordered DOS. The smearing effect of the disorder reduces the gap between oxygen-*p*-derived bands and the rest of the bands, leading to significant mixing between the oxygen-*p*-dominated states and the states having contributions from Fe t_{2g} states in the spin-up channel appearing in energy approximately between -4 eV to -2 eV, while the gap between the oxygen-derived states and the hybrid states arising from an admixture of Fe t_{2g}, Mo t_{2g}, and oxygen p crossing the Fermi level in the spin-down channel reduces to about 0.8 eV in configuration 1 and collapses in configuration 3, compared to a gap of about 2.5 eV in the case of the ordered sample (see Fig. 2). In the spin-up channel, disordering fills up the gap between crystal-field-split Fe t_{2g} and Fe e_g states observed in the ordered DOS, which are still below the Fermi energy, as well as the gap between the filled Fe-d states and the sharp density of states arising due to narrow Mo t_{2g} bands lying above E_F. We notice an overall broadening effect due to the disorder with the density of states reduced compared to that of the ordered calculation, arising from a spread in the site-diagonal energies induced by different Madelung potentials at the crystallographically inequivalent sites. The electronic structure in the disordered configurations is also affected by the off-diagonal disorder arising from changes in the cation neighbors and a consequent lowering of the symmetry. It is important to note that there is only a moderate polarization of the states at and near E_F in all the disordered configurations, in sharp contrast to the 100% polarization in the ordered sample. Since the transport mechanism involves charge carriers within a small energy range of E_F, the absence of a significant spin polarization of the states near E_F should have a pronounced effect on the low-field CMR properties of such disordered systems. It is interesting to note that experimentally there is indeed a complete suppression of the remarkable low-field CMR effect in the disordered samples.[5]

It should be kept in mind that the supercell calculations are actually ordered calculations that make use of the translational symmetry, which is not the case of a truly disordered material. Furthermore, even though we have used reasonably large supercells with four formula units containing 40 atoms, it is still a small number and therefore the various configurations that we can probe are limited. In particular, if a system experimentally approaches a chemically phase-separated scenario with only Fe ions somewhere spatially separated from the region with only Mo ions, such situations cannot be described within the present size of the supercell. It is possible that experimentally such phase-separated scenarios are actually achieved in certain extreme cases. We have probed the limit where disorder is still microscopic and does not lead to a larger length-scale phase separation.[14] In this context it will be ideal to perform a truly disordered calculation that is capable of probing local environment effects.[15]

In conclusion, we have performed electronic structure calculations for ordered and disordered Sr_2FeMoO_6 with positional disorder at the Fe/Mo sites. The effect of disorder has been modeled by supercell calculations. Disordering is found to destroy the half-metallic nature of the ordered compound; moreover, disorder leads to a significant reduction in the net magnetic moment. The reduction in the magnetic moment is caused by the decrease of the individual magnetic moments of different inequivalent Fe sites due to the various distributions of the neighbors and consequent bandwidth/correlation effects. Contrary to the result of a recent Monte Carlo study,[10] even the near-neighbor Fe-Fe and Mo-Mo interactions continue to be ferromagnetic also in the disordered configurations with an antiferromagnetic coupling between the Fe and Mo sublattices.

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