

## Electronic and magnetic properties of the double perovskite $\text{Sr}_2\text{FeMo}_x\text{W}_{1-x}\text{O}_6$

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### Abstract

The ordered double perovskite  $\text{Sr}_2\text{FeMo}_x\text{W}_{1-x}\text{O}_6$  compounds exhibit a transition from a ferromagnetic metal to an antiferromagnetic insulator with decreasing doping  $x$ . To understand the origin of this transition, using a double exchange type model with interaction between localized Fe spins and conduction electrons together with a tight-binding Hamiltonian and the renormalized perturbation expansion, we study the ferromagnetic phase and the ferro-antiferromagnetic transition with  $x$ .

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Recently, the double-perovskite oxide family has become very attractive in view of the remarkable low-field magnetoresistance properties up to room temperature of  $\text{Sr}_2\text{FeMoO}_6$  [1,2]. Different members of this family present very different electronic and magnetic properties.  $\text{Sr}_2\text{FeMoO}_6$  is in a half-metallic ferromagnetic state (complete polarization of the conduction electrons) with a high transition temperature ( $\sim 450$  K) [1,3]. On the other hand,  $\text{Sr}_2\text{FeWO}_6$  is known as an antiferromagnetic (AF) insulator with  $T_N \sim 37$  K [4]. As expected, a metal-insulator and magnetic transition has been reported as function of  $x$  in the substituted compounds  $\text{Sr}_2\text{FeMo}_x\text{W}_{1-x}\text{O}_6$  [5,6].

We consider a fully ordered perovskite structure in which Fe and Mo(W) occupy two interpenetrating sublattices in a rock-salt structure. Although most of the Fe-Mo samples present Fe-Mo disorder [1,2] indicated by a saturation magnetization lower than the ideal value  $4 \mu_B$  expected in a fully polarized ground state, it has been shown that the ordering increases with

the W concentration [6]. To picture these systems we follow the electronic scheme used recently to account for the effect of disorder [7]. Due to strong Hund's coupling Fe- $3d^5$  configuration is considered as a localized  $S = \frac{5}{2}$  maximum spin. Beside these local spins one has  $n = 1$  itinerant electrons coming from Mo (W) that can hop onto Fe sites only if its spin is antiparallel to the local spin. This scheme is consistent with the ferromagnetic ground state in which the conduction electrons couple antiferromagnetically to the local spins. To give a simple theoretical picture we neglect the correlations among the three possible  $t_{2g}$  orbitals and reduce the itinerant orbitals to one on each site. The orbital degeneracy of  $t_{2g}$  orbitals is accounted for at the end. According to the above considerations we write the model Hamiltonian for the ordered compound as

$$H = E_{\text{Fe}} \sum_{i \in \{\text{Fe}\}, \sigma} a_{i\sigma}^+ a_{i\sigma} + E_{\text{Mo}} \sum_{i \in \{\text{Mo}\}, \sigma} b_{i\sigma}^+ b_{i\sigma} - \sum_{\langle ij \rangle, \sigma} t_{\mu_i \sigma_j} (a_{i\sigma}^+ b_{j\sigma} + h.c.), \quad (1)$$

where  $E_{\text{Fe}}$  and  $E_{\text{Mo}}$  are the diagonal energies at the Fe and Mo sites respectively. The operators  $a_{i\sigma}^+$ ,  $a_{i\sigma}$  ( $b_{i\sigma}^+$ ,  $b_{i\sigma}$ )

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create and destroy an itinerant electron with spin  $\sigma$  at site  $i$  occupied by Fe (Mo), respectively. The hopping term  $t_{\mu,\sigma_j}$  is  $t$  when the localized spin  $\mu$  (+ for up and – for down) at site  $i$  is opposite to the itinerant spin  $\sigma$  ( $\uparrow$  or  $\downarrow$ ) and the hopping is zero when  $\mu$  and  $\sigma$  are parallel, this mechanism precludes the possibility to put an itinerant electron in the Fe sites with the same spin as the localized spin. The Mo–Mo hopping is ignored. The density of states for itinerant electrons is obtained from the local Green's functions for different sites in an interpenetrating Bethe lattice, using the renormalization perturbation expansion (RPE) method. We take the limit of infinite coordination number in the Bethe lattice ( $z \rightarrow \infty$ ). In this case  $zt^2$  scales as  $w^2/4$ ,  $w$  being half of the band width, and the Green's functions reduce to those used in a dynamical mean field approach

$$G_{\uparrow}^{\text{Mo}} = \frac{1}{\omega - E_{\text{Mo}} - (w^2/4)v_- G_{\uparrow-}^{\text{Fe}}} \quad (2)$$

$$G_{\uparrow-}^{\text{Fe}} = \frac{1}{\omega - E_{\text{Fe}} - x(w^2/4)G_{\uparrow}^{\text{Mo}}} \quad (3)$$

and similarly for down-spin electrons replacing  $v_-$  by  $v_+$ , where  $x$  indicates the concentration of Mo and  $v_{\pm} = (1 \pm m)/2$ ,  $m$  being the magnetization.

The relative position of Mo (W) and Fe itinerant states is a key parameter determining the Mo and Fe valence. For  $\text{Sr}_2\text{FeMoO}_6$  recent experiments converge towards an intermediate valence state  $\text{Fe}^{2.5+}$  [8,9], meaning that  $E_{\text{Mo}} \approx E_{\text{Fe}}$ . To keep the model applicable to other compounds we take  $E_{\text{Fe}}$  and  $E_{\text{Mo}} = \Delta$ . The valences of  $\text{Fe}(\text{Fe}^{2+})$  and  $\text{W}(\text{W}^{6+})$  in  $\text{Sr}_2\text{FeWO}_6$  [4] correspond to pushing to very high energy the relevant W d-state. The conduction electrons cannot hop to the W sites thus giving the factor  $x$  in Eq. (3).

In the ferromagnetic state the density of states consists of two bands with spectral weight  $x$  each and Fe localized states with weight  $(1-x)$  at  $\omega = 0$ . We find that the Curie temperature  $T_c \rightarrow 0$  with doping around  $x \sim 0.2$  (Fig. 1), which indicates the possibility of a magnetic transition. We see that this critical value of  $x$  is almost independent of  $\Delta$ . Therefore an antiferromagnetic super-exchange interaction between the Fe localized spins may stabilize an AF phase as in pure  $\text{Sr}_2\text{FeWO}_6$ . The magnetic structure has been reported recently [10]. Each Mo has an equal number of up- and down nearest-neighbors Fe-spins,  $z/2$  in our approach. This gives the same Green's functions for the AF state as for the paramagnetic state (Eq. 2 for  $v_{\pm} = \frac{1}{2}$ ). The difference of the electronic energy between the ferro and AF phases is shown in Fig. 2. The magnetic energy  $E_M/w$  resulting from the Fe–Fe superexchange interaction may be estimated as  $E_M \sim -T_N$ ,  $T_N$  being the Néel temperature. Estimating  $E_M/w \approx 0.008$  from our calculated value of  $T_c/w \approx 0.1$  for  $\Delta = 0$ , this pushes the critical concentration for the ferro–AF transition

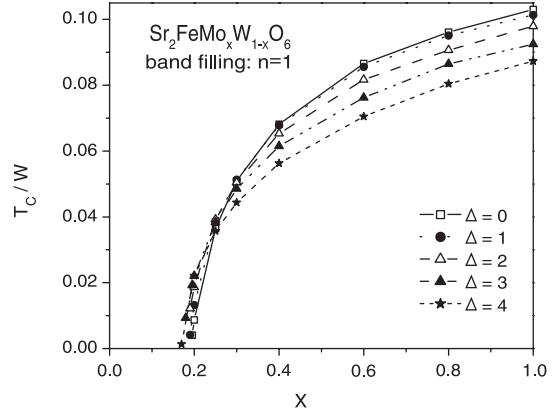


Fig. 1. Variation of the Curie temperature  $T_c$  as a function of the Mo concentration for different values of  $\Delta$ .

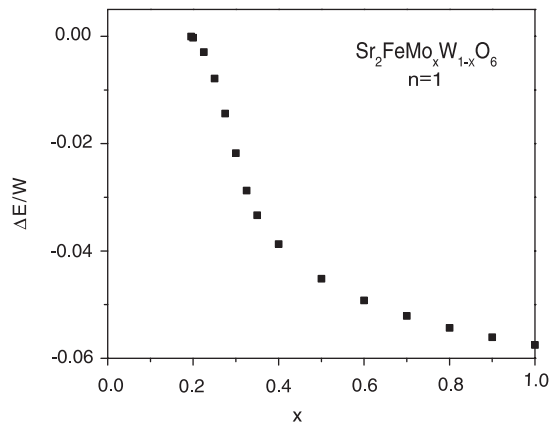


Fig. 2. Energy difference ( $\Delta E = E_F - E_{\text{AF}}$ ) for the ferro-antiferro phases vs. the Mo concentration.

towards  $x_c \sim 0.25$ , a value very near the experimental critical concentration.

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