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Non-collinear magnetic ordering in $R_{1/2}D_{1/2}MnO_3$ manganites

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Abstract

We discuss the possibility of a non-collinear magnetic structure to be the ground state of the half-doped manganites, as a natural consequence of the recently proposed alternative model to Mn^{3+}/Mn^{4+} charge ordering in such systems (Phys. Rev. Lett. 89 (2002) 097205). We show that the new non-collinear model and the collinear arrangement known as the CE-state, are almost two degenerate solutions for the refinement of the neutron powder diffraction studies that we have performed on half-doped manganites.

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The half-doped manganites $R_{1/2}Ca_{1/2}MnO_3$ (R: trivalent ion like Bi, La, Pr, Sm, Y) magnetic ground state is described by a collinear antiferromagnetic (AF) arrangement, known as the CE-type structure [1]. It results from superexchange (SE) interactions obeying the so-called Goodenough–Kanamori–Anderson (GKA) rules [2], and constructed assuming the spatial charge ordering (CO) of the Mn^{3+} and Mn^{4+} ions concomitant with the orbital ordering (OO) onto the Jahn–Teller (JT) active Mn^{3+} ions [3]. Beyond the Mn^{3+}/Mn^{4+} CO picture, we proposed that the paramagnetic CO phase is built from the ordering of what we have called two-manganese Zener polarons (ZP) [4,5].

Such a state was recently theoretically reproduced using unrestricted Hartree–Fock calculations [6], revealing in particular that the strong ferromagnetic (F) coupling within a Mn–O–Mn trio forming the ZP comes from the localization of the doped holes on the bridging oxygen. Besides this, all the Mn atoms become orbitally ordered Mn^{3+} ions: applying the GKA rules, this builds a different exchange topology than the original SE description, where half of the F–SE interactions are not satisfied by the reported collinear CE arrangement [1]

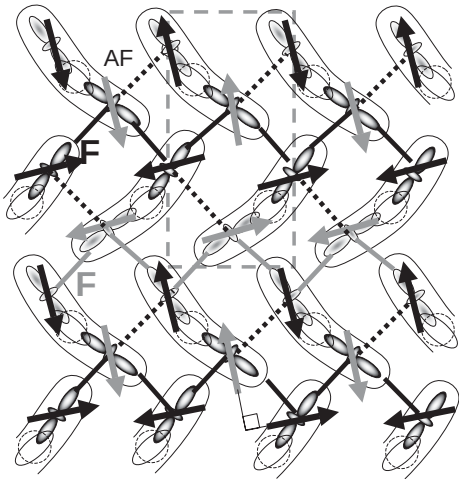

Fig. 1. The SE topology, in the ZP ordering scheme: arrows shows the non-collinear magnetic order. From one plane to the neighboring one, the OO is the same, while the spins are reversed.

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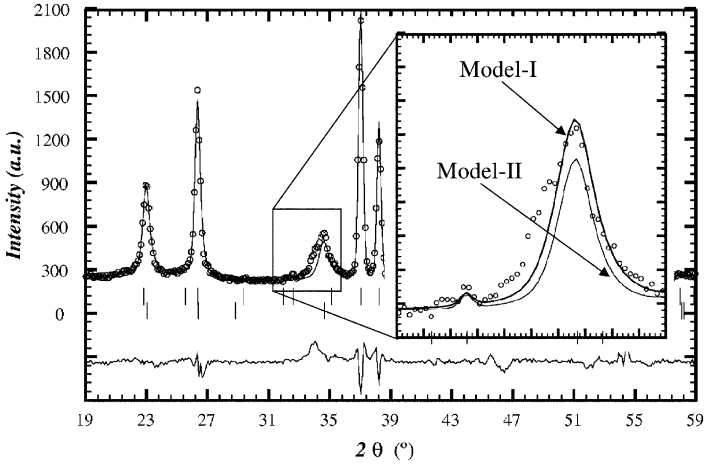

Fig. 2. Part of the NPD diagram of $Y_{1/2}Ca_{1/2}MnO_3$ at T = 1.5 K. Data (circles), refinement (solid line) and difference curve (bottom solid line) plotted for model II. Inset comparison of the refinements plots results with model I and II, in the region where a splitted magnetic peak is evidenced.

(see Fig. 1). For this reason, we have looked for alternative magnetic arrangements, which could be the ground state of this new exchange topology: one possibility is the non-collinear structure depicted in the Fig. 1, rendering all the F–SE coupling equivalent.

We have then compared the refinements of the collinear [1], and the non-collinear structures, denoted as model I and II, respectively, against the neutron powder diffraction (NPD) data collected on a prototypical $Y_{1/2}Ca_{1/2}MnO_3$ CO powder on the G41 powder diffractometer ($\lambda = 2.43$ Å) at the LLB in France. In model I, we refine two distinct values for the magnetic moments on the two distinct Mn sublattices, but only one moment value is refined in model II, that we consider as a constant moment structure. Only one additional parameter is refined, which is an angle allowing the moments to rotate in the $(\mathbf{a}, \mathbf{b})$ plane for both models, keeping the collinear arrangement in model I, and the orthogonal arrangement between two F-coupled ZP in model II (see Fig. 1).

We find a slightly better agreement for model I ($\chi^2 = 9.36\%$, $R_{Bragg} = 1.80\%$, $R_{Mag} = 6.14\%$), but let us stress why we believe that this fact alone is not sufficient to discard model II ($\chi^2 = 10.5\%$, $R_{Bragg} = 1.96\%$, $R_{Mag} = 9.71\%$). First, the CE state is known to present a characteristic short coherence length revealed on the data, by a selective broadening of the magnetic Bragg peaks: using the ionic CO framework, a specific modeling of this effect was proposed in Ref. [3]. We used this particular broadening model in all the refinements that we presented here. However, the attempts that we have made, performing refinements with alternative peak broadening laws, allowed us to obtain better agreements for both models, casting some doubts about the accuracy of the peak broadening law proposed in Ref. [3]. More importantly and whatever the peak broadening model, we assume a single magnetic phase in all our refinements, a fact that is clearly not justified since we evidence a splitted magnetic Bragg peak for which we do not have any explanation. This is a noteworthy feature, that we further suggest to be generic in all half-doped CO manganites since we have independently observed the same behavior in other $Pr_{1/2}Ca_{1/2}MnO_3$ and $Nd_{1/2}Ca_{1/2}MnO_3$ powders. Model I models better this splitted peak by an artificial broadening (see Fig. 2), but model II captures with an equal success than model I all the other magnetic intensities.

Hence, we argue that unless a satisfactory peak profile modeling is not achieved first, we cannot discriminate in between the two solutions: as a result of this analysis, the proposed non-collinear magnetic order for the half-doped manganites cannot be ruled out to be the possible ground state of this system.

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