H5. SPIN-WAVE DISPERSION IN ORBITALLY ORDERED $La_{0.5}Sr_{1.5}MnO_4$

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The colossal magneto-resistivity in manganites is only partially explained by the Zener double-exchange mechanism; the larger part of it appears to arise from the competition of two states: the metallic ferromag-netically ordered state on the one side and the insulating one with a cooperative ordering of charges, orbitals and spins (COS) on the other side [1, 2]. The insulator-to-metal transition consists in switching from a phase with long or short-range COS correlations into the metallic state where spins are aligned either by an external field or by spontaneous magnetic order. The combined COS ordering has first been studied in the pioneer work by Wollan and Koehler [3] and by Goodenough [4] proposing the so-called CE-type arrangement, which is illustrated in Fig. 1(*a*). For half doping, i.e. equal amounts of Mn^{3+} and Mn^{4+}, there is a checkerboard arrangement of different charges. In addition the e_g orbitals at the Mn^{3+} sites form zigzag chains. The CE-type charge and orbital arrangement will yield a ferromagnetic interaction in the zigzag chains and an [antiferromagnetic] interaction in-between.

The magnetic excitations in the ferromagnetic metallic manganites have been studied for many different compositions (for a recent summary see Ref. [5]). In view of the large amount of data on the ferromagnetic phases, it may be surprising that there is still no detailed study of magnetic excitations in the antiferromagnetic COS states. Besides the intrinsic complexity of the CE-type magnetic ordering, such a study is severely hampered by the twinning of the manganite crystals in the perovskite phases. We, therefore, have chosen the layered material $La_{0.5}Sr_{1.5}MnO_4$ to study the magnon dispersion in the COS state (Fig. 1).

We were able to separate the magnon branches parallel and perpendicular to the zigzag-chains, as only one twin orientation contributes to a given quarter-indexed magnetic superstructure reflection. When going from the *antiferromagnetic* zone center (0.75,-0.75,0) along the [1,1,0] direction one determines the spin-wave dispersion parallel to the zigzag chains (Fig. 2, right) and, when going along the [1-10] direction, one measures the dispersion perpendicular to the chains. This behavior is corroborated by the structure factor calculations presented in Fig. 2 as discussed below. The raw-data scans unambiguously demonstrate that

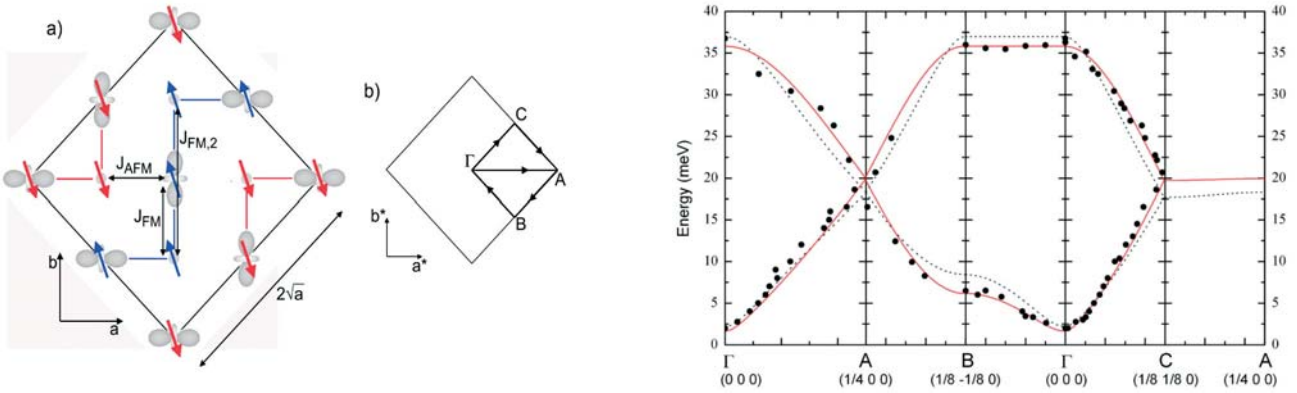

Figure 1. (a) Schematic representation of the CE-type ordering in the (*a,b*) plane of half-doped manganites with three magnetic interactions parameters. Notice that the FM zigzag chains run along the [110] direction. (*b*) Sketch of the magnetic Brillouin zone, displaying the high-symmetry points Γ = (0, 0, 0), *A* = (1/4, 0, 0), *B* = (1/8, -1/8, 0), *C* = (1/8, 1/8, 0) and the path of the calculated dispersion. (*c*) Dispersion of the magnetic excitations in $La_{0.5}Sr_{1.5}MnO_4$ in a direction parallel to [100] (Γ–*A*), perpendicular to the chains (Γ–*B*) and parallel to the chains (Γ–*C*). The solid and broken lines give the spin-wave dispersion calculated with a two parameter set.

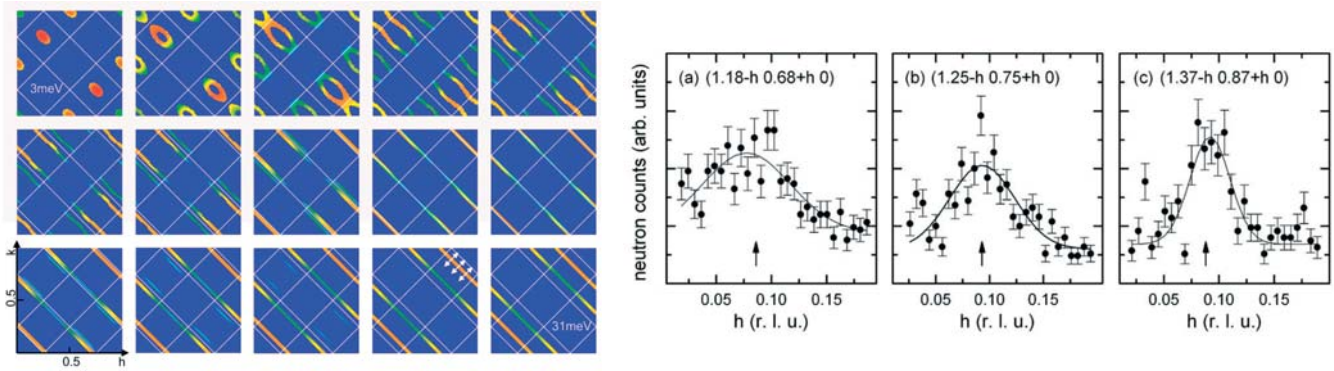

Figure 2. (left panel): Constant energy cuts through the calculated spin-wave structure factor with constant energy resolution of 2 meV within each plot, and with energy steps of 2 meV between adjacent plots, showing the dispersion and scattering distribution of the lowest magnon bands. (right panel): Constant energy scans along the direction indicated by arrows in the 29 meV cut (above) to experimentally verify the one-dimensional character of the high-energy magnetic scattering. The arrows indicate the expected positions of the magnon.

the dispersion along the zigzag chains is much steeper than perpendicular to them. The magnetic structure has to be considered as a weak antiferromagnetic coupling of strongly coupled ferromagnetic zigzag chains. The obtained magnon dispersion is presented in Fig. 1(*c*). The branch propagating along the chains (Γ–*C* path), is much steeper than the branch propagating perpendicular to it, (Γ–*B* path). At the magnetic zone boundaries *C* and *B* we find magnon energies of 19meV and 6.5 meV, respectively. At the C point where *q* is parallel to the chains, the end-point of the acoustic branch coincides with that of the lowest optic branch, whereas there is a large gap between these branches along the Γ–*B* path. The magnon branch along the [100] direction (Γ–A path, at 45° with respect to the chains), exhibits an intermediate dispersion.

The spin-wave dispersion has been calculated using the Holstein-Primakoff transformation with a simple spin-only Hamiltonian illustrated in Fig. 1(*a*). The Mn^{3+} and Mn^{4+} spins were fixed to the values $S = 2$ and $S = 1.5$, respectively. Taking into account only the two nearest-neighbor Mn^{3+}–Mn^{4+} spin interactions for pairs within and in-between the zigzag chains, J_{FM} and J_{AFM}, one obtains a good description of the measured dispersion denoted by broken lines in Fig. 1(*c*). However, there remain significant discrepancies: it is impossible to simultaneously describe the large initial slope of the spin-wave dispersion along the chains and the relatively lower zone-boundary frequencies. This behavior implies the relevance of an additional longer-distance interaction parameter acting along the ferromagnetic chains. Indeed, a fully satisfactory description is obtained by including a ferromagnetic interaction for Mn^{4+}–Mn^{4+} spin pairs connected through a Mn^{3+} site within a zigzag chain. Fig. 2 (left) presents the calculated magnon scattering intensities in the form of constant energy cuts. One can see how the anisotropic spin-wave cones develop around the magnetic Bragg peaks with finite structure factor. At intermediate energies those magnetic Brillouin zones in which there is no elastic scattering also contribute. Fig. 2 further illustrates that, well above the maximum of the acoustic magnon perpendicular to the zigzag chains, the system looks like a magnetically one-dimensional system as the magnons disperse only along the zigzag chains. The one-dimensional character was verified by special constant-energy scans (Fig. 2). The dominant ferromagnetic coupling is further seen in experiments upon heating across the charge and orbital ordering.

The strong ferromagnetic interaction in the zigzag chains not being restricted to the nearest neighbors indicates that electrons are not fully localized in the COS phase as well. The large and non-local ferro-magnetic interactions in the zigzag chains yield considerable resemblance between the COS and the metallic phases. This resemblance might be essential to understand the capability of manganites to switch between the metallic ferromagnetic and the insulating COS phases. These results have been published in reference [6].

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