CONFINED SPIN WAVES REVEAL AN ASSEMBLY OF NANOSIZE DOMAINS IN $La_{1-x}Ca_xMnO_3$ (x=0.17, 0.2)

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This study concerns the physics of metal-insulator transition in manganites with colossal magneto-resistance (CMR). This topics still provides a challenge for theories. The possibility of mixed-phases to explain the CMR properties in $La_{1-x}Ca_xMnO_3$ at hole doping x~0.3, has been pointed out by numerical studies [1]. However, in this doping range, the experiments are not conclusive. We have investigated two compounds at x=0.17 and x=0.2, close to x=0.22 where the ferromagnetic metallic (FM) phase with CMR properties occurs [2]. The compounds, which are ferromagnetic (F), exhibit a "quasi-metallic" behaviour below T_C (T_C=175K and 180K for x=0.17 and 0.2 resp.) as expected from double-exchange coupling. At lower temperature, however, they become insulating, which reveals additional processes not accounted by double-exchange. Inelastic neutron scattering performed in the quasi-metallic state, reveals that the spin-wave spectrum is strongly anomalous. Instead of a continuous cosine law as for a F spin wave dispersion (cf the dashed line in Fig. 1), nearly dispersionless levels are observed at large q values. They can be quantitatively described by spin waves confined inside finite-size clusters [3,4]. This analysis allows us to determine (i) the shape (evolving from two-dimensional for x=0.17 to three-dimensional for x=0.2), (ii) the size (decreasing from 4 to 2 lattice spacings with increasing x) and (iii) the super-exchange (SE) coupling constants inside the clusters. The use of SE coupling constants, in an Heisenberg model, indicates that the clusters are "hole-poor" or orbital-ordered, implying hole-rich and orbital-disordered boundaries. These observations could be crucial for clarifying CMR. Moreover, confined spin waves inside ultra-fine domains are observed for the first time.

Let us first focus on the x=0.2 compound, the results on x=0.17 being summarized below. Fig 1 displays the spin-wave spectrum in the quasi-metallic state (T=150K, left panel) and in the insulating state (right panel) along the [100], [010],[001] axis, superimposed because of twinning, in cubic indexation. At 150K, at small q up to q~0.25, the spectrum consists of a dispersed curve. Beyond, the intensity is divided into three levels moderately

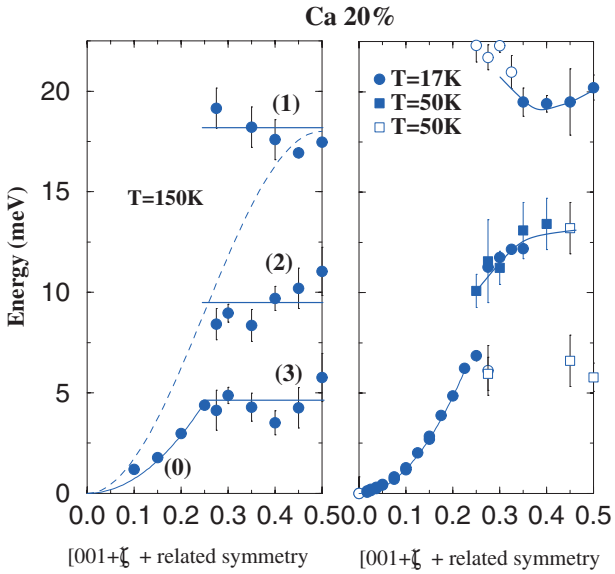

Figure 1. Spin wave dispersion along [001]+[010]+[001] in a zero-field experiment (after applying a magnetic field) at 150K (left panel) and 50K and 17K (right panel).

dispersed, numbered from (1) to (3). At low temperature, in the insulating state, the dispersion of the levels increases and the low-energy level (3) disappears. The same type of dispersionless modes are observed along all symmetry directions. The spin-wave spectrum along the [110],[011],[101] directions (diagonal of the cubic face) and along [111] (main diagonal of the cube for which all domains are equivalent) are reported in Fig. 2 at T=150K. Beyond a dispersed curve at small q values, only two levels are observed along [111] whereas several can be determined along [110].

From the expression for the energy of a propagating spin wave in a Heisenberg model, one gets relations between the zone boundary energies E_B, along each symmetry direction, and the superexchange (SE) magnetic coupling constants, $J_{a,b}$ along the **a** or **b** axis (equivalent) and J_c along the **c** axis of the perovskite cube. These relations are also valid for finite-size clusters, where E_B is the largest energy level. According to predictions [3], the lowest energy confined wave corresponds to a half-wave and its energy should be a sub-multiple of E_B. The cluster size ξ is deduced from

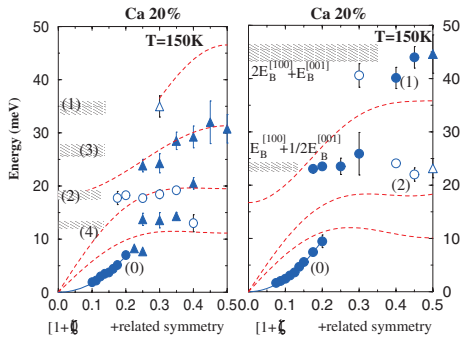

Figure 2. Magnetic excitations for q along [110],[101],[011] (left panel) and [111] (right panel) at T=150K, measured with unpolarised neutrons (circles) and polarized neutrons (triangles). Full (empty) symbols refer to modes with main (weak) intensity. The black hatched area correspond to calculated levels (see text) and the red dashed lines represent the phonon branches.

the ratio between this lowest energy level and E_B. In Figure 1, the largest energy level at E_B=18 meV and the level at 9 meV, are attributed to a full-wave and half-wave along [100] or [010], whereas the same level at 9 meV (accidental coincidence) and that at 4.5 meV are attributed to the full and half waves along [001] (see drawing in Fig 4). This assignment defines $J_{a,b}$ and J_c (reported in Fig 3) and the values of all energy levels along the other three symmetry directions (indicated by hatched areas, instead of lines, due to the experimental uncertainty), labeled (1) and (2) for [110], (3) and (4) for [011]+[101], in Fig 2-left panel, and (1) and (2) for [111] in Fig 2-right panel. The good agreement with experiment supports this attribution. A cluster size ξ of 2 lattice spacings is obtained. The small-q dispersed curve implies that the small domains are ferromagnetically coupled together.

A similar study for x=0.17 where the flat levels exist only in the **(a, b)** plane and not along the **c** direction, leads to the $J_{a,b}$ value reported in Fig 3 and determines a cluster size of 4 lattice spacings within the **(a, b)** plane. The general evolution of $J_{a,b}$ and J_c deduced from the dispersionless levels for x>0.125 (for x=0.17, J_c is obtained from E_B^{110} and E_B^{111}) and from the "normal" spin wave branch for x<0.125 (in the canted antiferromagnetic insulating state or CAFI) are reported in Fig 3. It shows a monotonic effect of the hole-doping on the SE coupling constants, starting from undoped $LaMnO_3$. These constants characterize an orbital-ordered or "hole-poor" spin system. A general evolution of the charge segregation with hole doping is displayed in Fig 4. The hole-rich domains, strongly anisotropic, embedded in an orbital-ordered matrix observed for x<0.125[5], percolate at x=0.125 (J_c=0), whereas small orbital-ordered domains with 2D (x=0.17) and 3D (x=0.2) character are formed as x increases beyond 0.125.

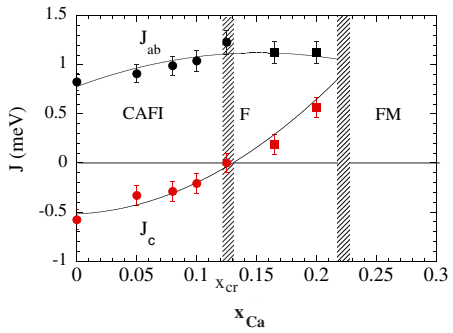

Figure 3. Variation of the magnetic coupling constants $J_{a,b}$ and J_c with doping. Vertical hatched lines indicate the CAFI/F and the F/FM transitions

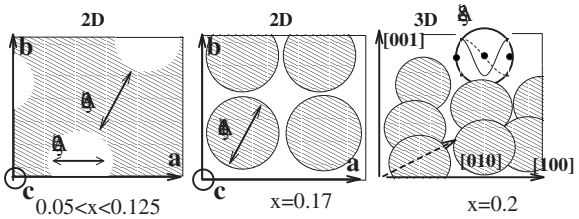

Figure 4. Schematic drawing showing the evolution of the charge segregation with doping x into 3 steps: from hole-rich (empty) clusters to hole-poor (hatched) domains with a 2D character and then to 3D isotropic clusters. A half and full standing waves are shown inside a cluster for x=0.2.

References

[1] A. Moreo, Seiji Yunoki, Elbio Dagotto, Science Vol. 283 pp 2034 (1999).
[2] P. Kober, Thesis Université Paris-Sud, defended on the 10th of December (2004)
[3] M. Hennion, F. Moussa, F. Wang, P. Lehouelleur, A. Ivanov, Y. M. Mukovskii and D. Shulyatev, Phys. Rev. Lett. (2005)
[4] P. V. Hendriksen, S. Linderoth and P. A. Lindgård Phys. Rev. B 48 pp 11600 (2000)
[5] G. Biotteau, M. Hennion, F. Moussa, J. Rodríguez-Carvajal, Pinsard and A. Revcolevschi, Y. M. Mukovskii and D. Shulyatev Phys. Rev. B 64 pp 104421 (2001)