Spin waves and metallic state of magnetoresistive manganites

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Abstract.
La1−x(B = Sr, Ca, Ba)xMnO3 manganites are known to undergo upon doping a transition from an insulating ferromagnetic state to a puzzling metallic ferromagnetic phase showing colossal magnetoresistance properties. Revisiting spin wave measurements in this metallic phase, we observe strong similarities with the insulating x ∼ 1/8 regime, suggesting that the same physics of small ferromagnetic domains is at play [1, 2]. We argue that all the anomalies in this metallic state could be explained in this inhomogeneous picture, in continuity with the x ∼ 1/8 range.

1. Introduction
The discovery of CMR in cubic manganites has aroused a lot of experimental and theoretical research interest for the last decade and a large number of studies have been devoted to the description of the evolution of spin dynamics in these materials [3, 4, 5]. Several anomalies have been identified in the spin wave spectra, which are suggested to be generic features of the unusual metallic state of CMR manganites [6, 7, 8]. As these unconventional spin waves remain unexplained, new experimental efforts are needed to extend our understanding of these compounds and of their puzzling properties. We report on measurements of spin waves in La1−x(B = Sr, Ca, Ba)xMnO3 samples, B = Sr or Ca, for dopings covering a large part of the phase diagram, from the quasimetallic and insulating parts with x(Sr) = 0.15 to the metallic region with x(Sr) = 0.175, 0.2, 0.3 and x(Ca) = 0.3. The measurements have been carried out at the Laboratoire Léon Brillouin (1T, 2T, 4F spectrometers) and at the Institut Laue Langevin (IN8 spectrometer). PG002 was used as analyser, together with Cu001, Cu002, Cu220 or PG002 as monochromators, yielding an energy resolution (FWHM) varying from 1.5 meV to 4 meV at intermediate energy transfer E = 16meV.

In all samples, we observe spin wave spectra consisting in a quadratic dispersed branch at the zone centre, characteristic of a three-dimensional (3D) ferromagnetic state, and nearly wave-vector independent levels at the zone boundary. In the metallic phase, the characteristic energies of these levels remain approximately constant, while their intensity shows a peculiar temperature and doping dependence jumping from the lower to the upper levels as T decreases or as x increases. Since both insulating and metallic regimes have similar spin wave spectra, we
speculate that the same physics is at play across the insulating-metallic transition, and that it might be an important ingredient for the CMR phenomenon.

2. The insulating $O'O''$ phase around $x \sim 1/8$

We first describe the spin dynamics in the insulating $x \sim 1/8$ phase of the phase diagram. Here, the spin wave spectrum for a doping level $x(Sr) = 0.15$ along $[1+q,0,0]$ is found to be very close to that observed for $x(Sr) = 0.125$ [1]. Below $T_C=230K$, a quadratic $E = Dq^2$ dispersion is observed for $q \leq 0.25$, as well as several nearly $q$-independent levels for $q > 0.25$. Below $T_{O'O''}=180K$, $D$ increases, and a gap opens at $q=0.125$. Concomitantly, the levels become $q$-modulated. The experiments allow to resolve three levels, lying at $E=7.5, 15, 23$ meV for $q=0.5$ (see Figure 1a). In addition, we note that along [111] and [100], the energies of the zone boundary modes with strongest intensity differ by a factor of $\approx 2$, which is expected for a 2D

Figure 1. a) Spin wave dispersion for $x = 0.15$ in the insulating phase, below $T_{O'O''}=180K$. Open and closed symbols correspond respectively to small and large observed neutron intensities. b) Sketch of the (a,b) plane, showing ferromagnetic domains, along with definition of the couplings $J_{OO}$ and $J_{inter}$. c) Calculated spin wave dispersion for $J_{inter} = J_{OO}/5$. d) Spin wave dispersion for $x(Sr) = 0.30$ (metallic phase). The dashed line shows the cosine law expected according to [11]. e) Energy scans performed at various temperatures at the zone boundary $Q = (1.5, 0, 0)$. The arrows denote the positions of the different levels. Note that the low energy ones are better observed at high temperature. f) Calculated spin wave dispersion in the extended version of the model (see text).
coupling.

In [1], we proposed a qualitative interpretation of these spectra in terms of quantized spin waves in $2D$ $4a \times 4a$ ferromagnetic domains (see Figure 1b). Here, we propose an improved description where the spin dynamics is calculated in the framework of a Heisenberg model with two NN interactions: $J_{OO}$ couples respectively Mn spins within the same domain while $J_{inter}$ couples spin across the domain boundaries. This kind of superstructure, also proposed in the context of high-$T_c$ superconductivity [10], implies that the holes lie, in average on the oxygen ($O$) atoms of the boundaries. Note that $x = 1/8$ precisely corresponds to half-filling, which may explain the stability of the $O'/O''$ phase. For $J_{inter} = 0$, the spin dynamics is that of quantized spin waves within $4a \times 4a$ domains. If $J_{inter} \leq J_{OO}$, the quantized spin waves in different clusters couple, and the levels become dispersive. Choosing $J_{inter} = J_{OO}/5$ allows a good description of the data, at least for $q \geq 0.15$, as shown in Figure 1c.

3. The metallic phase

On entering the metallic phase, we observe strong similarities with the $x \sim 1/8$ range. Again, the spin wave spectra consist of two components: a quadratic regime near the zone center and several levels close to the zone boundary (Figure 1d). Figure 1e shows representative raw data taken at $Q = (1.5,0,0)$ for 298K, 250K, 150K and 14K in the $x(Sr) = 0.3$ compound ($T_C=370K$). The two levels at 15 and 22 meV typical of the $x=1/8$ range, are properly resolved together with new ones lying at $E \approx 32$ meV, $\approx 41$ meV and $\approx 51$ meV. As the temperature decreases, the intensity progressively shifts from the two low-energy levels towards the higher energy ones. This evolution can be correlated with the concomitant increase of the stiffness constant $D$. Similar results are observed for other dopings within the metallic phase, namely for $x(Sr) = 0.175$ ($T_C=270K$), $x(Sr) = 0.2$ ($T_C=325K$), and $x(Ca) = 0.3$ ($T_C=255K$). This temperature dependence is thus a general trend of the metallic phase.

These observations can be understood in an extended version of the above mentioned model. We now assume disordered $2D$ $F$ domains of $4a \times 4a$ size, embedded in a $3D$ $F$ matrix and take into account the ferromagnetic $NN$ coupling $J_{OD}$, acting between spins within this $3D$ medium. Basically, $J_{OD}$ has the same physical meaning as $D$ ($J_{OD}=D/2a^2$) induced by hopping electrons. A qualitative agreement between experiment and numerical simulations is obtained if $J_{OD} \gtrless J_{OO}$. Figure 1f shows an example of calculated spin wave spectrum along [100] performed with a density of clusters of 50%, $J_{OD} = 1.75 J_{OO}$ ($J_{OO}=1.7$ meV) and $J_{inter}=J_{OO}/5$. In these calculations, the levels are reminiscent of quantized spin waves within the domains, while the hybridization with the $3D$ medium blurs the large gaps at low $q$ and adds new levels at higher energy.

This interpretation provides also a simple explanation for the so-called "softening" effect. As shown by the dashed lines on Figure 1d, the cosine law expected from conventional double-exchange model[11], defined from the $q \equiv 0$ dispersion, yields a zone boundary energy value clearly above the experimental one. Previous studies have proposed that this anomaly may be due to fourth neighbor coupling term [6, 7, 8], assuming the existence of a single branch. Our observations, however, unveil domain excitations caracterised by $J_{OO}$. As $J_{OD} \gtrsim J_{OO}$, this causes an apparent "softening" of the zone boundary energy.

We finally discuss the spin dynamics, aiming at a global understanding from the undoped or slightly doped to the metallic region of the phase diagram. To this end, Figure 2 shows the evolution of the different coupling constants as a function of doping. We start from zero doping and lightly doped samples, characterized by A-type magnetic structure. The AF coupling along the c-axis progressively disappears and reaches $J_c(AF) = 0$ at about $x=12.5\%$, corresponding to the onset of the ferromagnetic structure in the $(x,T)$ phase diagram (see [8]). Concomitantly, the (planar) ferromagnetic coupling $J_{a,b}(F)$ very slightly increases. For larger dopings, the system breaks up in an assembly of weakly connected 4x4 domains where the spins are ferromagnetically
Figure 2. Evolution with doping of the various coupling constants describing the spin dynamics in manganites. $J_{a,b}(F)$ and $J_{c}(AF)$ correspond to the ferromagnetic and antiferromagnetic exchange observed in the undoped and slightly doped compounds. $J_{OO}$ and $J_{OD}$ are described in the text.

coupled via $J_{OO}$. It is worth noting that $J_{a,b}(F)$ is nothing but the precursor of $J_{OO}$. We also report the evolution of $J_{OD}$, which is reminiscent of the itinerant charge carriers. In the low doping range, it corresponds to the additional spin wave branch with a small gap (the anisotropy is not shown here) originally discussed in [12]. Reflecting the growing number of holes introduced in the system, $J_{OD}$ progressively increases with doping. Finally, we observe that, in the insulating phase, $J_{OD} \leq J_{A,b}(F)$, whereas the O’O” charge ordered phase corresponds to $J_{OD} = J_{OO}$, and the metallic phase to $J_{OD} \geq J_{OO}$.

4. Conclusion

In conclusion, our results show that the metallic state of manganites can be described in terms of quantized spin waves in 2D domains embedded in a 3D matrix. The remarkable similarities of the spin dynamics observed in several compounds, indicate that these inhomogeneities have the same origin whatever $T$, $x$ or the average cation size. We speculate that these domains might be an important ingredient for the CMR phenomenon.

References

[1] M. Hennion et al., Phys. Rev. B 73 104453 (2006)
[2] S. Petit et al., Phys. Rev. Lett. 102 207201 (2009)
[3] Nanoscale Phase Separation and Colossal Magnetoresistance, edited by E. Dagotto (Springer-Verlag, Berlin, 2002)
[4] Y. Motome and N. Furukawa, Phys. Rev. B 71 014446 (2005)
[5] Sanjeev Kumar et al., Phys. Rev. Lett. 97 176403 (2006)
[6] Y. Endoh et al., Phys. Rev. Lett. 94, 017206 (2005)
[7] F. Ye et al., Phys. Rev. Lett. 96, 047204 (2006) and Phys. Rev. B 75 144408 (2007)
[8] F. Moussa et al., Phys. Rev. B 76 064403 (2007).
[9] H. Y. Hwang et al., Phys. Rev. Lett. 75 914 (1995)
[10] Boris Fine Cond-mat/0404488
[11] N. Furukawa, J. Phys. Soc. Jpn 65 1174 (1996)
[12] M. Hennion et al, Phys. Rev. B 56 R497 (1997)