PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

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Magnetic dynamics in colossal magnetoresistive perovskite manganites

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We have measured the spin-wave dispersion throughout the Brillouin zone of the double-exchange ferromagnet $La_{0.7}Pb_{0.3}MnO_3$ by using inelastic neutron scattering. An unexpectedly simple Heisenberg Hamiltonian with solely a nearest-neighbour coupling accounts for the entire dispersion relation. The value of the Curie temperature T_C calculated from the derived coupling constant agrees with the experimental value to within 15%. The results, including damping of the zone boundary spin waves near T_C, can be fully understood in the framework of the double-exchange Hamiltonian. In the paramagnetic phase of the two-dimensional analogue $La_{1.2}Sr_{1.8}Mn_2O_7$, we have observed long-lived antiferromagnetic clusters coexisting with ferromagnetic fluctuations. This effect must be included in theories that attempt to explain giant magnetoresistance in manganites, at least in two dimensions.

Keywords: spin waves; antiferromagnetic fluctuations; critical magnetic scattering; Heisenberg Hamiltonian

1. Introduction

Over the past few years there has been a huge revival (Kusters *et al*. 1989; von Helmolt *et al*. 1995; Jin *et al*. 1994; Asamitsu *et al*. 1995) of interest in doped, rare earth manganese perovskites, $RE_{1-x}A_xMnO_3$ (RE = La, Nd, Pr; A = Ca, Sr, Ba, Pb) because of the colossal magnetoresistance that they exhibit. The parent compounds are antiferromagnetic insulators, such as $LaMnO_3$, into which holes can be introduced by substitution of the RE^{3+} cation with an A^{2+} cation. With sufficient doping the materials can become ferromagnetic metals with Curie temperatures $T_C \approx 100$–400 K. As the transition temperature is approached from below, there is a large decrease in the electrical conductivity, σ, and the materials remain poor conductors in the paramagnetic phase. The electrical conductivity appears to depend only on the temperature and applied-field-dependent magnetization M, so that near T_C, where the magnetic susceptibility is large, σ is very sensitive to the applied field.

The bulk magnetic properties can be qualitatively explained by the double-exchange mechanism (Zener 1951; Anderson & Hasegawa 1955; de Gennes 1960). In the doped material all Mn sites have a local spin $S = 3/2$ in localized T_{2g} orbitals, and a fraction $(1-x)$ have an extra electron in the higher energy E_g orbitals, which

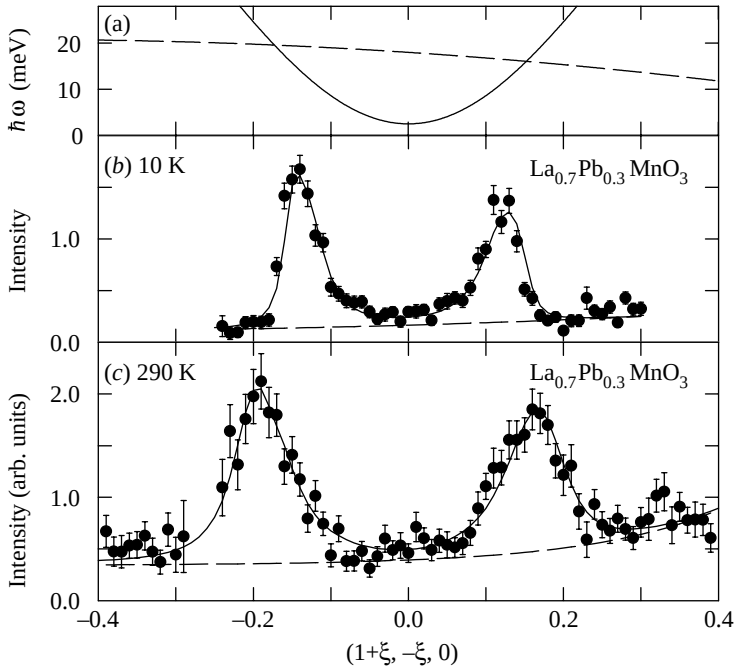

Figure 1. Nearly constant energy transfer scan along $(\xi, -\xi, 0)$ through the (1,0,0) reciprocal lattice point. (*a*) $\hbar\omega$ as a function of ξ for the scan (dashed line) and spin-wave dispersion relation (solid line). (*b*) and (*c*) show scattering intensity for 10 K and 290 K respectively. Solid lines are fits as described in text. (After Perring *et al.* (1996).)

are hybridized with the oxygen p-orbitals to form a narrow conduction band. The carrier electrons can hop, without loss of spin polarization, to adjacent Mn sites with empty E_g orbitals. Strong intrasite exchange aligns the carrier spins parallel to the local spins, making it energetically favourable to hop between Mn sites with parallel local spins. This accounts for both the ferromagnetic exchange coupling and carrier mobility that is sensitive to the applied field near T_C. The model successfully explains the doping dependence of T_C (Furukawa 1995*a*) and σ as a function of M when the two quantities are expressed as fractions of zero-magnetization conductivity and saturation magnetization (Tokura *et al.* 1994; Furukawa 1995*b*), but local lattice distortions due to strong electron–phonon coupling have been proposed (Millis *et al.* 1995, 1996; Röder *et al.* 1996) as an essential ingredient, necessary to explain the absolute value of T_C as well as the absolute conductivity and its activated temperature dependence in the paramagnetic phase. An important question is to what extent can the double-exchange model be used as a starting point to describe the magnetism in the perovskites?

We have performed inelastic neutron-scattering experiments to investigate the effective magnetic Hamiltonian in a 2 g single crystal of the colossal magnetoresistance compound $La_{0.7}Pb_{0.3}MnO_3$ (Perring *et al.* 1996). Thequestions we address are the following. (*a*) Is there a simple effective spin Hamiltonian? (*b*) What is the magnitude of the exchange constants, and can they explain the Curie temperature? (*c*) How well does the double-exchange model explain the spin dynamics? We have also looked for any memory of the undoped parent's antiferromagnetism in the spin

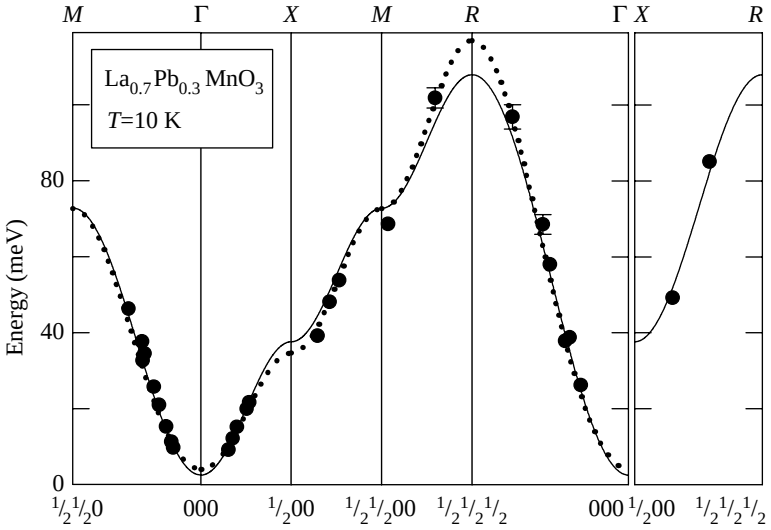

Figure 2. Spin-wave dispersion relation along all major symmetry directions. Points correspond to data at 10 K, lines to the best fit to the dispersion relation for Heisenberg ferromagnet with nearest-neighbour exchange only (solid line), double-exchange ferromagnet (dotted line, from Furukawa (1996)). Fit parameters as in main text. (After Perring *et al.* (1996).)

dynamics of a two-dimensional analogue which also exhibits colossal magnetoresistance (Perring *et al*. 1997).

Most of the experiments were performed on the time-of-flight spectrometer HET at the ISIS pulsed spallation neutron source. The incident beam energy E_{i} can be fixed at any desired value and the times of arrival of scattered neutrons in a large multi-detector array are recorded. The detectors in any one bank probe momentum transfers $\boldsymbol{Q}$ in a plane in reciprocal space, so that several spin-wave peaks along symmetry directions can be measured with one setting of E_{i} and the crystal orientation. Figure 1*b*, *c* shows example spin waves near the Brillouin zone centre at $0.03T_{\mathrm{C}}$ and $0.82T_{\mathrm{C}}$. The solid lines in figure 1*b*, *c* represent the convolution of the instrumental resolution with the theoretical cross-section for a Heisenberg ferromagnet with nearest-neighbour exchange (NNHFM), further convolved with a damped simple harmonic oscillator to allow for finite spin-wave lifetimes due, for example, to decays into electron–hole pairs.

The dispersion relation at 10 K, obtained from just a few E_{i} and crystal settings, is shown in figure 2, showing an overall spin-wave bandwidth of approximately 100 meV. Figure 3*a* shows conventional time-of-flight scans for two fixed detector angles, which demonstrate that at 10 K the spin waves are well defined even near the (0.5,0.5,0.5) zone boundary point, implying that there is little damping of the spin waves through interactions with the carriers.

A general model for the effective spin Hamiltonian is that for the Heisenberg ferromagnet (HFM), with couplings J_{ij} between pairs of spins at sites $\boldsymbol{R}_i$ and $\boldsymbol{R}_j$, $H = -\sum_{ij} J_{ij}\boldsymbol{S}_i \cdot \boldsymbol{S}_j$. Surprisingly, we find that a single-exchange constant which couples only nearest-neighbour spins, $2JS = 8.8 \pm 0.2$ meV, together with a small value for the gap, $\Delta = 2.5 \pm 0.5$ meV, is sufficient to account for the entire dispersion relation in figure 2. The value of the gap is probably not significant because it is much

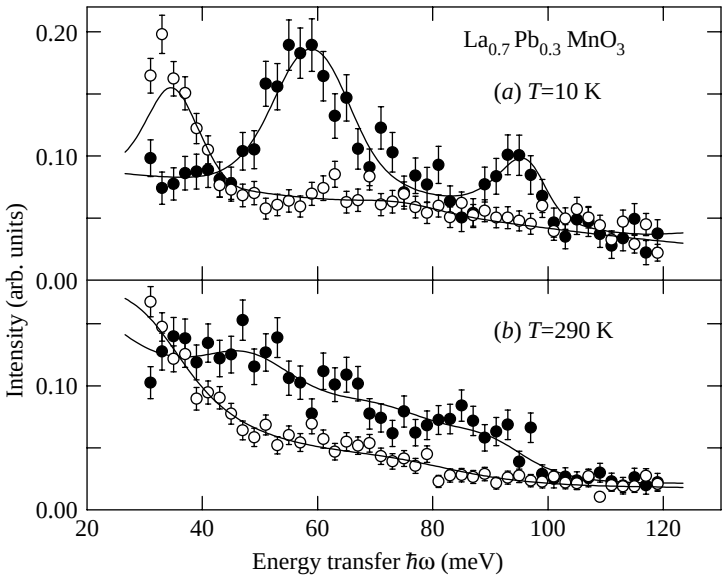

Figure 3. High-energy scans for $E_{\mathrm{i}} = 200$ meV in $La_{0.7}Pb_{0.3}MnO_3$ at (*a*) 10 K and (*b*) 290 K. Scattering angle ranges are $\phi = 9.3$–10.6° (solid circles), which scans almost along $(1/2, 1/2, 1/2+\xi)$, and $\phi = 15.6$–17.4° (open circles), which scans well away from $(1/2, 1/2, 1/2)$ and so provides a good estimate of non-magnetic scattering. Solid lines are simultaneous fits to the two angular ranges with the model as in figure 1 (described in text). (After Perring *et al.* (1996).)

smaller than the lowest energy spin-wave energies measured, and consequently is sensitive to precise details of the model to the instrument resolution function. Addition of second-nearest-neighbour and third-nearest-neighbour coupling constants does not improve the quality of the fit, and the values of $2JS$ and Δ only alter by amounts similar to the error bars on those quantities. The T_{C} for the NNHFM, including the effect of fluctuations, is given by $k_{\mathrm{B}}T_{\mathrm{C}}/J = 2.90S(S+1) - 0.36$ (Rushbrooke *et al.* 1974, eqn 5.4). Taking $S = 3/2 + (1-x)/2 = 1.85$ and using our fitted value for $2JS$ the expression for T_{C} yields 410 K, only 15% greater than the experimental value of 355 K.

The double-exchange Hamiltonian can be written as

$$H = -t \sum_{\langle i,j\rangle,\sigma} (c^*_{i\sigma} c_{j\sigma} + \mathrm{h.c.}) - \frac{J_{\mathrm{H}}}{S} \sum_i \boldsymbol{S}_i \cdot \boldsymbol{\sigma}_i, \tag{1.1}$$

where t is the carrier hopping integral and J_{H} is the intrasite exchange (Hund's rule) coupling between the electrons and local spins $\boldsymbol{S}_i$. A calculation of the spin waves for this Hamiltonian (Furukawa 1996) shows that in the infinite coupling limit ($J_{\mathrm{H}} \to \infty$) the dispersion relation reduces to precisely that for the NNHFM, with the spin-wave bandwidth determined by t. For $x = 0.3$ the best fit is with $t = 0.18$ eV. A somewhat better fit to the data is obtained with $J_{\mathrm{H}}/t = 12$, with $J_{\mathrm{H}} = 3.2$ eV, $t = 0.26$ eV (corresponding to a half-carrier bandwidth $W = 1.6$ eV). The values of both the bandwidth and the intrasite exchange are realistic values without any renormalization. The T_{C} for the Hamiltonian in equation (1.1) has been calculated for the case of classical spins in infinite dimensions (Furukawa 1995*a*). For the fitted

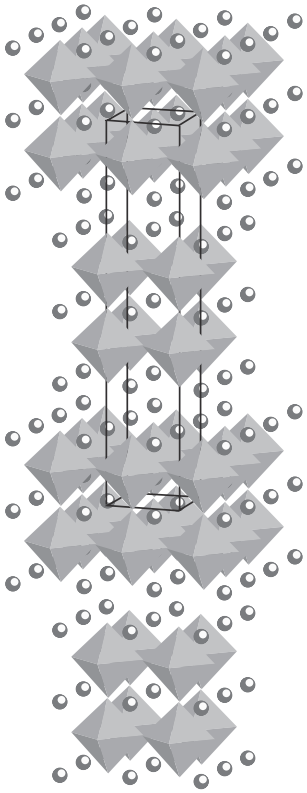

Figure 4. Structure of $La_{2-2x}Sr_{1+2x}Mn_2O_7$. The Mn ions are at the centre of the MnO_6 octahedra. Circles denote La and Sr.

values of J_H and t with $x = 0.3$ the estimate is $T_C = 390$ K, again close to the experimental value of 355 K.

Raising the temperature to 290 K $\equiv 0.82T_C$ has a dramatic effect on the high-energy spin waves. Figure 3*b* shows that near the zone boundary the spin waves broaden almost to the point of disappearing. The inverse lifetime γ increases from 10 ± 2 meV at 10 K to 27 ± 5 meV at 290 K. On the other hand, the lower-energy spin waves shown in figure 1*c* remain well defined with γ increasing only to 5 meV. The double-exchange mechanism provides a qualitative explanation of the broadening (N. Furukawa, personal communication). At low temperatures all carriers have their spins aligned 'up' and the energy cost to flip the spin is about $J_H S \sim O$ (eV). This energy corresponds to the minimum Stoner excitation energy, which is in turn well above the maximum spin-wave energy, and so the spin waves remain well defined throughout the zone. Near T_C there are local spins pointing in all directions so a significant fraction of the carriers at the Fermi energy have spin 'down'. The spins of the carriers therefore can be flipped with little energy cost and so can readily couple to the spin waves and dampen them.

Our experiments have shown that the spin dynamics, at least in the ferromagnetic phase, are well described by the double-exchange Hamiltonian. At 355 K the value of T_C in $La_{0.7}Pb_{0.3}MnO_3$ is near the maximum value observed in $RE_{1-x}A_xMnO_3$. It is interesting to compare our results with recent spin-wave measurements in the

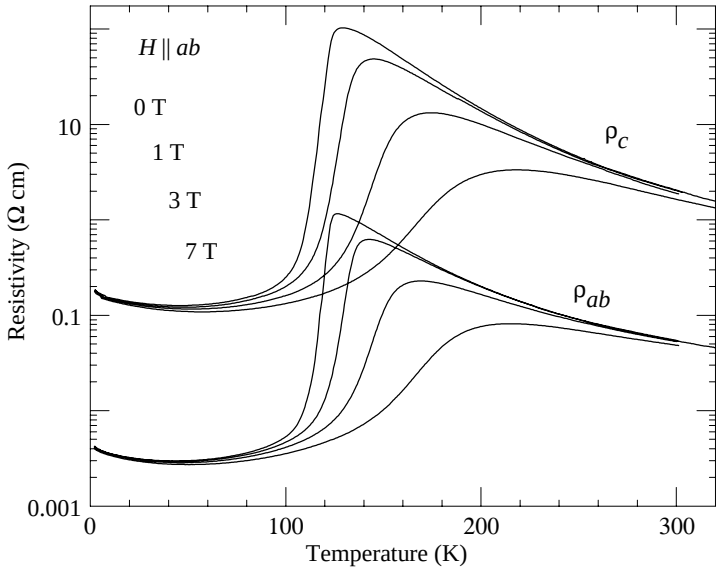

Figure 5. Temperature dependence of the resistivity of $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.4$) in the basal plane, ρ_{ab}, and along the c-axis, ρ_c.

Table 1. *Spin wave stiffnesses in various manganites with hole doping* $x \approx 0.3$

compound and hole doping	T_C (K)	spin wave stiffness D (meV Å^2)	spin wave energy at $(\frac{1}{2}\,0\,0)$ zone boundary (meV)
[a]$La_{0.7}Sr_{0.3}MnO_3$ ($x = 0.30$)	378	188 ± 8	—
[b]$La_{0.7}Pb_{0.3}MnO_3$ ($x = 0.30$)	355	133 ± 4	37
[c]$Pr_{0.67}Sr_{0.33}MnO_3$ ($x = 0.33$)	301	165	20
[d]$La_{0.67}Ca_{0.33}MnO_3$ ($x = 0.33$)	250	170	—
[c]$Nd_{0.7}Sr_{0.3}MnO_3$ ($x = 0.30$)	198	165	∼20

[a]Martin *et al*. 1996. D derived from fit to spin waves with energies not greater than 30 meV along $(\xi\,\xi\,0)$.
[b]Perring *et al*. 1996. D derived from fit to spin waves throughout Brillouin zone.
[c]Fernandez-Baca *et al*. 1998. D derived from fit to spin wave energies not greater than 3 meV.
[d]Lynn *et al*. 1996. D from spin wave energies not greater than 1 meV.

lower T_C materials $Pr_{0.63}Sr_{0.37}MnO_3$ ($T_C = 301$ K) and $Nd_{0.7}Sr_{0.3}MnO_3$ ($T_C = 198$ K) (Hwang *et al*. 1998; Fernandez-Baca *et al*. 1997), for which measurements are restricted to momentum transfers between the zone centre and the (0.5,0,0) zone boundary point (see table 1). In both materials well-defined spin waves exist to the zone boundary, but with a marked softening of the dispersion relation as (0.5,0,0) is approached that cannot be reproduced by the spin-wave dispersion derived from the Hamiltonian of equation (1.1) (Furukawa 1996; Hwang *et al*. 1998). Close to T_C in the ferromagnetic phase the spin waves are damped in $Pr_{0.63}Sr_{0.37}MnO_3$, just as in $La_{0.7}Pb_{0.3}MnO_3$, but in the low T_C compound $Nd_{0.7}Sr_{0.3}MnO_3$ intense quasi-elastic scattering appears, which has been attributed to the presence of magnetic polarons (de Teresa *et al*. 1997). The picture that emerges is one where the double-

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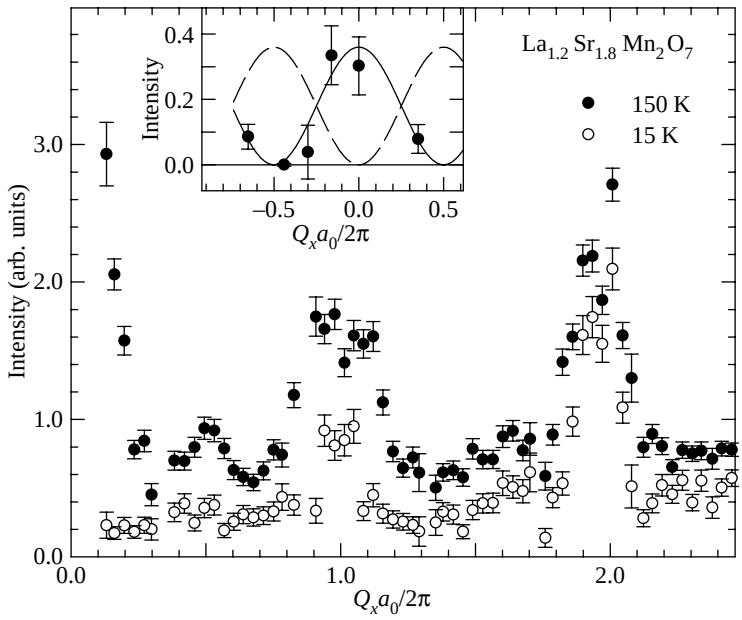

Figure 6. Momentum dependence of scattering integrated between 1 meV and 2 meV for h along $(h, 0, Q_z c_0/2\pi)$. Inset shows dependence on Q_z in $(0.5, 0, Q_z c_0/2\pi)$. (After Perring *et al.* (1997).)

exchange model alone becomes progressively unable to explain the spin dynamics as T_C is reduced. Softening of the spin waves near the zone boundary may be due to slow fluctuations towards doubled nuclear unit cells (Hwang *et al.* 1998). This picture is consistent with theoretical work (Millis *et al.* 1995, 1996; Röder *et al.* 1996) which emphasizes the important role played by electron–lattice coupling through a dynamic Jahn–Teller effect in the explanation of the magnetotransport, for which experimental data show that the magnitude of the magnetoresistance becomes more enhanced in compounds with lower T_C (Hwang *et al.* 1995).

Because spin fluctuations are enhanced for lower dimensional magnets, we have examined the fluctuations in the paramagnetic phase of a two-dimensional analogue of $La_{0.7}Pb_{0.3}MnO_3$. The material is $La_{1.2}Sr_{1.8}Mn_2O_7$ (Moritomo *et al.* 1996) (T_C = 126 K, with $x = 0.4$ of the Mn sites doped with holes), made of MnO_2 bilayers equivalent to double-layer slabs cut from $LaMnO_3$ perpendicular to [0 0 1], spaced by layers of $(La,Sr)_2O_2$, which separate the slabs both electrically and magnetically (figure 4). It is tetragonal, with lattice parameters $a_0 = 3.87$ Å in the basal plane (≡Mn–O–Mn distance within the bilayers) and $c_0 = 20.14$ Å. The transport data show the same qualitative behaviour as in the pseudo-cubic perovskites $RE_{1-x}A_xMnO_3$, but the resistivity perpendicular to the bilayers is two orders of magnitude higher than within the bilayers (figure 5).

Figure 6 shows the integrated scattering intensity between 1–2 meV as a function of h in $(h, 0, Q_z c_0/2\pi)$ for $Q_z a_0 \ll \pi$ at 15 K and 150 K = $T_C + 24$ K. In addition to the ferromagnetic critical scattering that appears just above T_C at $\boldsymbol{Q}$ near nuclear zone centres (0,0,0), (1,0,0) and (2,0,0), there is also weaker scattering near the zone boundary point (0.5,0,0). Near (1.5,0,0) it is very much weaker, clearly showing the magnetic, rather than structural, origin of the scattering. The inset shows the Q_z

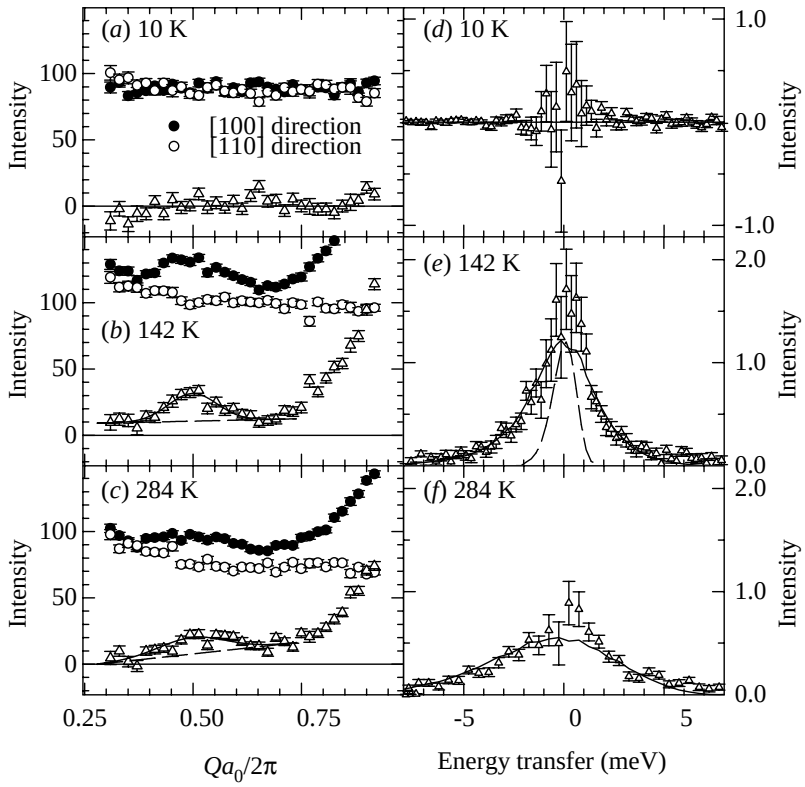

Figure 7. (*a*)–(*c*) Momentum dependence of scattering integrated between −5 and +5 meV along [1 0 0] (closed circles) and [1 1 0] (open circles). In addition to scattering at (0.5,0,0), warming above T_C induces ferromagnetic critical scattering visible along (1,0,0) but not visible along (1,1,0) in the Q range shown. This is because the (1,1,0) reciprocal lattice point is at $Q = 1.414 \times (2\pi/a_0)$. If there were any scattering centred on (0.5,0.5,0) it would peak at $Q = 0.707 \times (2\pi/a_0)$. Open triangles represent the difference between the two directions. The solid lines show fits to Gaussians on a linear background (dashed line). (*d*)–(*f*) show the analogous differences in the scattering integrated over momentum between 0.39 and $0.63 \times (2\pi/a_0)$ and plotted as a function of energy transfer. The dashed line in (*e*) represents the instrumental resolution, the solid lines are the fits to the model described in the text. (After Perring *et al.* (1997).)

dependence in $(0.5, 0, Q_z c_0/2\pi)$ integrated between the same energy limits. The data are consistent with the variation (solid line) expected for ferromagnetic alignment of the spins in adjacent planes in the bilayers, but inconsistent with $\sin^2(Q_z a_0/2)$, the variation expected for antiferromagnetic alignment (dashed line). Within the basal plane, there is a peak at a position equivalent to $(0.5, 0, Q_z c_0/2\pi)$ just above T_C, but not at $(0.5, 0.5, Q_z c_0/2\pi)$ (figure 7*b*). Together, the results imply that the magnetic unit cell is doubled along only one axis parallel to the Mn–Mn direction in the basal plane, with the spins ferromagnetically correlated in the perpendicular direction in the basal plane and with the spins in the immediately adjacent plane.

Figure 8 shows how the intensity of the correlations develops with increasing temperature. As the ordering temperature is approached from below, the antiferromagnetic correlations appear only very close to T_C, but persist to at least 284 K = $2.3T_C$,

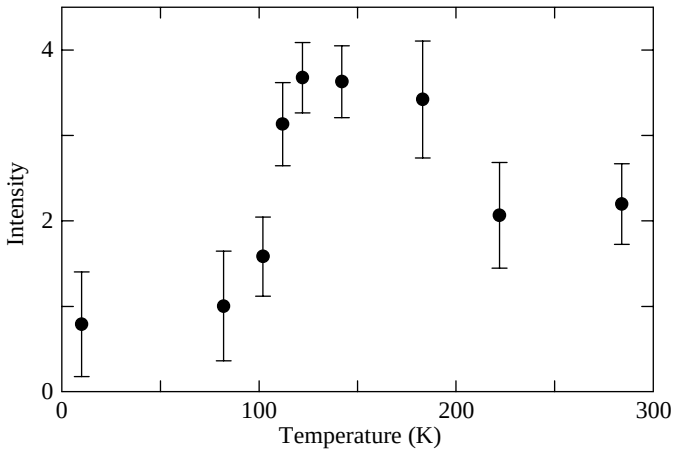

Figure 8. Intensity of the antiferromagnetic correlations integrated over the energy range −5 to +5 meV and the momentum range 0.39 and 0.63 × $(2\pi/a_0)$ along [1 0 0].

the highest temperature measured. Fitting a Gaussian line-shape to the momentum dependence of the fluctuations in figure 7*b*, *c* shows that the correlation length ξ decreases from $2.4 \pm 0.3a_0$ to $1.7 \pm 0.6a_0$ when the temperature is raised from $1.13T_C$ to $2.3T_C$. The lifetime of the correlations can be estimated by fitting the energy dependence in figure 7*e*, *f* to the product of a quasi-elastic Lorentzian, with momentum dependence Gaussian in the plane of the bilayers and $\cos^2(Q_z a_0/2)$ perpendicular to the bilayers. The result is that although the inverse lifetime increases over the same temperature range, from 1.7 ± 0.2 meV to 5.5 ± 1.7 meV, it remains substantially less than the thermal energy $k_B T = 24.5$ meV.

The coexistence of short-range slowly fluctuating antiferromagnetic correlations with ferromagnetic critical scattering in the paramagnetic region may have implications for the mechanism of the magnetoresistance in $La_{1.2}Sr_{1.8}Mn_2O_7$. The result shows that the carriers are moving in a medium which contains both ferromagnetic and antiferromagnetic Mn–Mn bonds. In the framework of the double-exchange model, antiferromagnetic bonds are a barrier to carrier motion, so that on frequency scales large compared to the fluctuations there is a localization mechanism. As temperature is increased the frequency scale of the fluctuations increases, partly destroying this mechanism. One explanation for our results suggests for the transport properties in the paramagnetic region that spins belong to fluctuating ferromagnetic clusters for most of the time, but are occasionally part of antiferromagnetic regions which can be viewed as moving walls between oppositely orientated ferromagnetic domains.

In conclusion, we have shown that in the pseudo-cubic perovskite $La_{0.7}Pb_{0.3}MnO_3$ the spin dynamics and Curie temperature are well explained by the double-exchange mechanism without requiring any additional physics. The evidence from compounds with lower values of T_C is that the double-exchange model alone is insufficient to explain the spin dynamics (Hwang *et al*. 1998; Fernandez-Baca *et al*. 1997). In a layered analogue of the pseudo-cubic perovskites we have observed antiferromagnetic fluctuations in the paramagnetic phase which may have implications for the explanation of the magnetoresistance at least in two dimensions.

References

Anderson, P. W. & Hasegawa, H. 1955 *Phys. Rev.* **100**, 675.

Asamitsu, A., Moritomo, Y., Tomioka, Y., Arima, T. & Tokura, Y. 1995 *Nature* **373**, 407.

de Gennes, P.-G. 1960 *Phys. Rev.* **118**, 141.

de Teresa, J. M., Ibarra, M. R., Algarabel, P. A., Ritter, C., Marquina, C., Blasco, J., Garcia, J., Delmoral, A. & Arnold, Z. 1997 *Nature* **386**, 256.

Fernandez-Baca, J. A., Dai, P., Hwang, H. Y., Kloc, C. & Cheong, S.-W. 1998 *Phys. Rev. Lett.* **80**, 4012.

Furukawa, N. 1995*a* *J. Phys. Soc. Jpn* **64**, 2754.

Furukawa, N. 1995*b* *J. Phys. Soc. Jpn* **64**, 2734.

Furukawa, N. 1996 *J. Phys. Soc. Jpn* **65**, 1174.

Hwang, H. Y., Cheong, S.-W., Radaelli, P. G. , Marezio, M. & Batlogg, B. 1995 *Phys. Rev. Lett.* **75**, 914.

Hwang, H. Y., Dai, P., Cheong, S.-W., Aeppli, G., Tennant, D. A. & Mook, H. A. 1998 *Phys. Rev. Lett.* **80**, 1316.

Jin, S., Tiefel, T. H., McCormack, M., Fastnacht, R. A., Ramesh, R. & Chen, L. H. 1994 *Science* **264**, 413.

Kusters, R. M., Singleton, J., Keen, D. A., McGreevy, R. & Hayes, W. 1989 *Physica* B **155**, 362.

Lynn, J. W., Erwin, R. W., Borchers, J. A., Huang, Q., Santoro, A., Peng, J.-L. & Li, Z. Y. 1996 *Phys. Rev. Lett.* **76**, 4046.

Martin, M. C. *et al*. 1996 *Phys. Rev.* B **53**, 14285.

Millis, A. J., Littlewood, P. B. & Shraiman, B. I. 1995 *Phys. Rev. Lett.* **74**, 5144.

Millis, A. J., Shraiman, B. I. & Mueller, R. 1996 *Phys. Rev. Lett.* **77**, 175.

Moritomo, Y., Asamitsu, A., Kuwahara, H. & Tokura, Y. 1996 *Nature* **380**, 141.

Perring, T. G., Aeppli, G., Hayden, S. M., Carter, S. A., Remeika, J. P. & Cheong, S.-W. 1996 *Phys. Rev. Lett.* **77**, 711.

Perring, T. G., Aeppli, G., Moritomo, Y. & Tokura, Y. 1997 *Phys. Rev. Lett.* **78**, 3197.

Röder, H., Zang, J. & Bishop, A. R. 1996 *Phys. Rev. Lett.* **76**, 1356.

Rushbrooke, G. S., Baker, G. A. & Wood, P. J. 1974 In *Phase transitions and critical phenomena* (ed. C. Domb & M. S. Green), vol. 3. New York: Academic.

Tokura, Y., Urushibara, A., Moritomo, Y., Arima, T., Asamitsu, A., Kido, G. & Furukawa, N. 1994 *J. Phys. Soc. Jpn* **63**, 3931.

von Helmolt, R., Wecker, J., Holzapfel, B., Schultz, L. & Samwer, K. 1993 *Phys. Rev. Lett.* **71**, 2331.

Zener, C. 1951 *Phys. Rev.* **82**, 403.

Discussion

T. Venkatesan (*University of Maryland, USA*). In the data for the magnetization for the Ruddlesden–Popper phases, Dr Perring did not show the full data, but if you look at the full temperature dependence for the $n = 2$ material, at about 300 K, you actually see a very small rise, a sort of ferromagnetic transition in the $n = 2$ data. Is he aware of that?

One of the key issues in this material is whether we have a two-dimensional ferromagnetism occurring around about 300 K, which is prevented from establishing a three-dimensional correlation. What you are seeing at low temperatures is really an overall three-dimensional correlation being established. What prevents this two-dimensional system from interacting and forming three-dimensional system at the

higher temperature? The antiferromagnetic fluctuations that he referred to may be one reason.

T. G. Perring. I haven't thought about that. Is Dr Venkatesan saying that above about 300 K, there is a jump in the magnetization? The experimental data are limited solely by the piece of equipment that was available in which to do it, but we now have more kit which can go to higher temperatures.

D. McK. Paul (*University of Warwick, UK*). Dr Perring has measured the excitations at low temperatures. Does he have any indication what the exchange is between these layers and within the layers? How two dimensional is it?

T. G. Perring. This is a plot of the data which assumes that the data are two dimensional in their character. If they are three dimensional, this is just going to be some completely filled in blur of intensity.

D. McK. Paul. But Dr Perring has got a bilayer, so it is a modulated object that he is going to see, not a single line of scattering.

T. G. Perring. Yes, and the modulation is going to give me a very approximate measure of what the exchange constant is between the bilayers. I actually tried to measure it in these materials, but I couldn't because of the mosaic of 3° on this crystal actually had the effect of washing it out. All I know is that it is small.

D. McK. Paul. Why not integrate along the rod to see what the q-dependent susceptibility does, in order to determine how two dimensional the system is?

G. A. Gehring (*University of Sheffield, UK*). Has Dr Perring measured the spin waves in the two-dimensional system over the whole two-dimensional Brillouin zone? He could then look for how well that fitted a Heisenberg model, and if there was evidence of the antiferromagnetic exchange coupling in the spin-wave dispersion relations.

T. G. Perring. The history of this experiment was a bit of serendipity; I set out precisely to measure the spin waves and got interested in the antiferromagnetic stuff which came in some other bank of detectors which I wasn't looking at. So as a result I haven't looked in any detail at the spin waves. This is the spin waves along the $(h\,0\,0)$ direction and the only thing that is apparent is that if you try and draw a line through here which is $\sin^2$-type variation from the Heisenberg ferromagnet, then it's not going to make it. It's much steeper near the $(1\,0\,0)$ point and the exchange constant which you would fit here for the zone boundary spin wave. So I think I can say qualitatively already that the Heisenberg local moment does not explain the spin waves in the basal plane of this material very well at all.

A. J. Millis (*The Johns Hopkins University, USA*). The Maryland NIST group, Jeff Lynn and co-workers, have, I think, published quite a lot of data on the LaCa materials in which they see in particular that the spin-wave stiffness at low q and low energy doesn't change very much from material to material within the LaCa family, whereas T_C and the magnetoresistance and all these things change quite substantially. It is my impression that the things which do change substantially are the zone boundary magnons which soften and broaden appreciably so that as you go away from the metallic ones you go away from the simple Heisenberg model quite

dramatically. This suggests that the La/Pb material discussed in this paper is very unlike most other manganites.

T. G. Perring. That's quite right, the various exchange constants have been measured. In fact for the lead-doped material is actually surprisingly soft compared to these other materials, including the NdSr material which has a T_C of 200 K. This is in part a function of the range over which you take your data, most of these data are being taken over a very restricted part of the Brillouin zone, the data on the Pd-doped are being taken throughout the Brillouin zone, so the sheer weight from points coming away from the zone centre is going to have some influence on the overall fit which I get for the data, but nevertheless they do look a little bit anomalous compared with the other materials.

A. J. Millis. The quantity D which is present in table 1 is defined in terms of the behaviour near the zone centre, and the differences between the materials appear at high q.

T. G. Perring. Absolutely. I'm saying that potentially this is a shortcoming of the fitting of the Pb-doped because it is not sensitive to exactly what is happening at the zone centre.

J. Pierre (*Laboratoire Louis Néel, Grenoble, France*). There are also experiments by the group at Saclay, Fermon and Rousseaux, on $LaMnO_3$, which is of course antiferromagnetic, but one can differentiate the antiferromagnetic exchange within the basal plane from the ferromagnetic exchange perpendicular to it. For doping with Ca they observe that the antiferromagnetic exchange is going diametrically down and that the ferromagnetic exchange increases as a function of doping.

J. M. D. Coey (*Trinity College, Dublin, Republic of Ireland*). One of the most interesting results obtained by the Saclay group (which they obtained in the 5% Ca-doped crystal) was the observation of a very soft branch which they tentatively associated with magnetic polarons. Has Dr Perring observed any such very soft magnetic excitations and does he agree with the explanation?

T. G. Perring. I'm not sure I agree with the explanation. I don't quite understand the results in fact. I have not observed any such results, but I have only been looking at the 30% doping and the nature of the experiment that I'm doing is not sensitive to the very low energies.

D. McK. Paul. The other thing which came from Jeff Lynn's experiment was a kind of diffusive mode which came in at low energy. This could be some kind of cluster in the system.

A. J. Millis. The central peak observed by Jeff Lynn is most pronounced in the samples with higher resistance and lower T_C, and as you change the materials' parameters and give them lower resistance and higher T_C, i.e. to make them look more like the LaPb material, the central peak goes away.

T. G. Perring. This is the central peak in the two-dimensional material, about 20 K below the Curie temperature, so possibly the same thing is going on.

S.-W. Cheong (*Bell Laboratories/Lucent Technologies, USA*). By now we have a lot of data about the central peak issue and the zone boundary modes, and we all

know the T_C is a very important parameter here, so when T_C is high and close to the optimum value like 370 K, then the spin-wave dispersion curve follows a simple Heisenberg model with nearest-neighbour interactions. When T_C is reduced from the optimum value, then the spin-wave stiffness does not change much as a function of reduced T_C; however, a diffusive component, the so-called central peak, develops at the zone boundary and increases with decreasing T_C. In addition, the energy of the zone boundary mode goes down and the magnon lifetime increases with decreasing T_C. Another general trend with decreasing T_C is that the spin-wave stiffness changes with temperature and becomes more like first order.

P. B. Littlewood (*University of Cambridge, UK*). Based on some of the things we've heard about lattice effects, one might expect to see them prominently in the data of coupled magnon–phonon effects. Does Dr Perring see any evidence for that kind of thing?

T. G. Perring. No I don't, and that is why the Pb-doped material seems to stand out as rather peculiar in that I don't see any such effects in the spin-wave data and I think it would be interesting to look for dynamical lattice effects.

A. J. Millis. Further to the previous remark, again I think the spin–lattice coupling must be the reason why the zone boundary magnons get extremely broad and come down in energy as you go away from the optimally doped T_C. As far as I know, nobody knows how to calculate it.

T. G. Perring. Kaplan & Mahanti (1997), in their calculations on a chain using the double-exchange model, only claim to see this effect, but it is a chain and not a three-dimensional model they work with.

A. J. Millis. That calculation should apply to the LaPb material and I'm sure that because there are only six sites, what they are seeing is just finite size effects.

D. Khomskii (*University of Groningen, The Netherlands*). If I understood correctly, this layered compound contains not $3^+/4^+$ Mn as in the cubic perovskite, but rather $2^+/3^+$. Isn't this perhaps relevant?

T. G. Perring. No, the valences are the same.

Additional references

Kaplan, T. A. & Mahanti, S. D. 1997 *J. Phys. Condens. Matter* **9**, L291.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES