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Physica B 385–386 (2006) 66–68

PHYSICA B

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Spin waves and phonons in the CMR ferromagnet $La_{0.70}Ca_{0.30}MnO_3$

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Abstract

The spin and lattice excitations in the CMR ferromagnet $La_{0.70}Ca_{0.30}MnO_3$ (LCMO30) have been studied by the use of polarized inelastic neutron scattering. This work has been carried out on a large single crystal using the IN20 polarized triple axis spectrometer at the Institut Laue Langevin, Grenoble, France. The spin waves and the lowest lying optical phonons both show a considerable degree of broadening and the use of polarized neutrons has been essential in order to separate these two components of the scattering.

PACS: 75.30.Ps; 75.47.Gk; 75.47.Lx

Keywords: CMR; Magnon–Phonon interaction

1. Introduction

The understanding of the colossal magnetoresistance (CMR) effect—the unusually large change in electrical resistance in response to a magnetic field—in certain materials is among the most interesting unresolved problems in condensed matter physics. It is known that the richness of the novel properties of these materials is related to the strong interplay between their charge, lattice, spin and orbital degrees of freedom [1]. The most studied of these materials are the doped perovskite manganites $A_{1-x}B_xMnO_3$ [where A is a trivalent ion (La^{3+}, Pr^{3+}, Nd^{3+}, etc) and B is a divalent ion (Ca^{2+} or Sr^{2+})] with $x \approx 0.3$. The basic microscopic mechanism responsible for the CMR effect is believed to be the double-exchange (DE) interaction, where ferromagnetism and electrical conductivity arise from hopping of the itinerant e_g electrons from trivalent Mn^{3+} to tetravalent Mn^{4+} sites. The physics of the CMR effect, however, is far from being completely understood. It is known, for example, that the DE mechanism alone cannot account for all the observed features of the spin dynamics of the CMR manganites. While the DE model predicts (in its lowest approximation) that spin-wave spectra of these systems should be similar to those for a nearest neighbour Heisenberg ferromagnet [2], various experiments have shown that this is not the case. What was observed instead, is that the spin waves are softened, broadened and dampened near the zone boundary [3–5]. We would like to remark, however, that these spin-wave measurements were performed using unpolarized neutrons and, while the wavevector and temperature dependence of the observed excitations seemed to indicate that they were magnetic in origin, it is essential to rule out any possible phonon contribution to the observed scattering. For this reason we have performed polarized neutron scattering experiments in order to separate the contributions of the phonons and the spin waves in

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doi:10.1016/j.physb.2006.05.104

$La_{0.70}Ca_{0.30}MnO_3$ (LCMO30). We have predominantly studied the behaviour in the $[0, 0, \xi]$ and $[\xi, \xi, 0]$ directions (using a pseudo-cubic cell for LCMO30) and in this paper we report the results for the $[\xi, \xi, 0]$ direction. The results for the $[0, 0, \xi]$ will be reported in a future publication along with supplementary (unpolarized) measurements of the phonons. For the $[\xi, \xi, 0]$ direction our polarized neutron measurements have revealed that most of the broadening reported from unpolarized neutron scattering experiments is due to the inability to separate the phonons from the scattering from spin waves.

2. Experimental

For this experiment we used the high-quality single crystal of LCMO30, grown by the floating zone technique, that was previously used in Ref. [4]. In the following we refer to wavevectors using the pseudocubic cell for LCMO with the lattice parameter $a \approx 3.86$ Å. Consequently, in this notation, the reciprocal lattice vectors are of the form $\mathbf{Q} = (H, K, L) = (q_x, q_y, q_z)2\pi/a$.

The polarized inelastic neutron experiments were performed using the IN20 polarized neutron triple axis spectrometer at the Institut Laue Langevin, Grenoble, France. In the measurements reported in this paper the spectrometer was operated in the half-polarized setup of Holden and Stirling [6]. This configuration makes use of a polarizing monochromator crystal and an unpolarized neutron analyzer, in combination with a horizontal magnetic field along the scattering wavevector **Q**. In this configuration the magnetic field constrains the neutron polarization **P** to be parallel or anti-parallel to the wavevector **Q**. When $\mathbf{P} \cdot \mathbf{Q} = 1$ the inelastic neutron cross section contains two terms corresponding to the nuclear (phonon) scattering plus twice the ferromagnetic spin waves [7]. When $\mathbf{P} \cdot \mathbf{Q} = -1$ only the nuclear (phonon) scattering contributes to the cross section and the scattering from the ferromagnetic spin excitations is eliminated.

The particular combination we used on IN20 was a Heusler (1 1 1) monochromator with fixed vertical and variable horizontal focusing, a pyrolitic graphite (0 0 2) analyzer with fixed vertical and horizontal focusing, and a 2 T horizontal magnetic field parallel to the wavevector **Q**. In order to vary the neutron polarization a Mezei spin flipper was used between the polarizing monochromator and the sample. This technique is very efficient method for the separation of phonon and spin-wave scattering from ferromagnets, provided that the horizontal field is large enough to align the ferromagnetic domains of the sample. For our measurements we verified that the 2 T applied field was more than sufficient to ensure this condition by performing some inelastic measurements at 15 K in a fully polarized (Heusler monochromator to Heusler analyzer) configuration and observing that while the spin wave cross section was observed in the (±) channel it was absent in the (∓) channel.

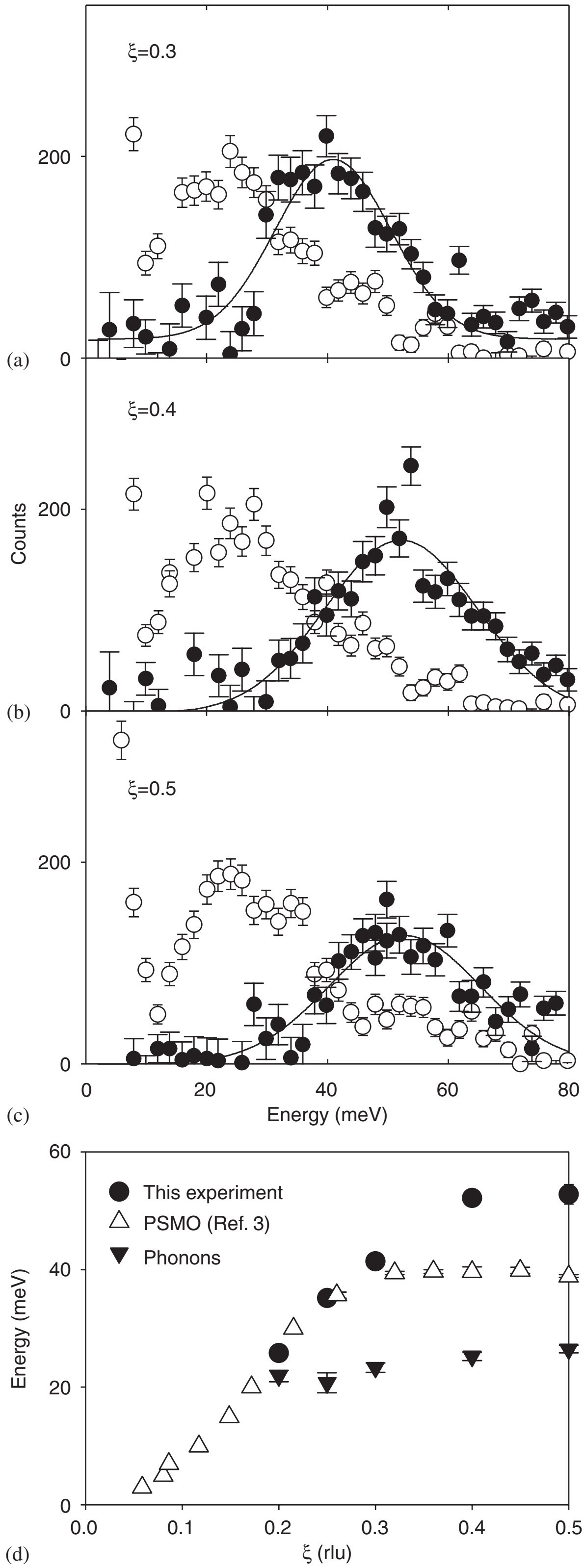

Fig. 1. (a)–(c). Scattering from phonons (open circles) and spin waves (full circles) in constant-Q scans $Q = (\xi, \xi, 2)$ for $\xi = 0.3$, 0.4, 0.5 rlu. (d) Spin wave dispersion relation along the $(\xi, \xi, 0)$. The full circles correspond to data from this polarized neutron experiment, the open triangles are from the unpolarized neutron experiment of Ref. [3] in PSMO. The solid triangles correspond to the centre of mass of the phonon scattering observed in the present experiment.

In our particular realization of the Holden–Stirling half-polarized setup the signal for the spin flipper OFF (FF) corresponds to purely nuclear (phonon) scattering, while the difference of the signals for flipper ON (FO) and OFF (FF), i.e. (FO–FF), corresponds to the ferromagnetic spin wave excitations. The measurements reported in this paper were performed with a fixed $K_F = 4.1\,\text{Å}^{-1}$ at a temperature $T = 15\,\text{K}$ along the $[\xi, \xi, 2]$ direction for $\xi = 0.2$, 0.25, 0.3, 0.4 and 0.5 with the spin flipper OFF (FF) and ON (FO) as explained above.

The panels (a)–(c) of Fig. 1 show the measured signal with the spin flipper OFF (FF, open circles) and the difference between the signals between the spin flipper ON and the spin flipper OFF (FO–FF, full symbols) for $\xi = 0.3$, 0.4 and 0.5. These correspond to the pure phonon and pure spin-wave scattering, respectively, and illustrate the power of this technique to separate these two different contributions to the scattering that cannot be resolved in an unpolarized neutron scattering experiment.

We note that the spin-wave energies near the zone boundary shown in Fig. 1(c) are higher than those reported in Ref. [3] for $Pr_{0.63}Sr_{0.37}MnO_3$ (PSMO) from unpolarized neutron measurements. This discrepancy can be explained by the fact that unpolarized neutrons can only measure the combined signals from the two contributions mentioned above. This is especially true at wavevectors close to the zone boundary where magnetic and lattice contributions to the scattering are both expected to be significant. In these cases the reported [4] positions of the spin wave peaks in fact correspond to the centroids of the phonon and spin wave peaks. To illustrate this point we have plotted in Fig. 1(d) the energies of the spin waves (filled symbols) and phonons (filled triangles) measured in this experiment. For comparison we also include the spin-wave dispersion relation of PSMO reported in Ref. [3] (open triangles). We can appreciate from this figure that the zone boundary energies from the unpolarized neutron experiment (open triangles) indeed approximately correspond to the centroid of the spin wave and phonon scattering reported in this paper.

Finally, we would like to point out that the spin wave and phonon peaks observed in this experiment are still significantly broader than the instrumental resolution. The observed broad phonon peaks are likely to be the superposition of the two lowest Σ_3 transverse optical phonon branches in this system [8].

3. Summary and discussion

We have performed a half-polarized neutron inelastic scattering experiment in the ferromagnetic phase of LCMO30 to separate the spin wave and phonon contributions to the excitation spectrum. From our measurements we conclude that, in the $[\xi, \xi, 0]$ the softening and broadening of the spin waves near the zone boundary is not as severe as reported from earlier unpolarized inelastic neutron scattering experiments [3]. The zone boundary energy for this branch is about 50 meV, which is still lower that the energy expected from the approximation to the Heisenberg Hamiltonian.

Acknowledgements

We thank the Institut Laue Langevin for the provision of neutron beam time to carry out this work. Oak Ridge National Laboratory is managed by UT-Battelle, LLC, for the US Department of Energy under contract DE-AC05-00OR22725. The work at UT was supported by the US NSF DMR-0453804 and DOE DE-FG02-05ER46202. The work at ANSTO was supported from project 0165P-6 and the Australian Major Research Facilities Programme.

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