

Composition Dependence of the Spin Wave Stiffness Parameter  
in  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  CMR Materials

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**Composition dependence of the spin wave stiffness parameter in  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  CMR materials**J.J. Rhyne<sup>a</sup>, H. Kaiser<sup>a</sup>, L. Stumpe<sup>a</sup>, J.F. Mitchell<sup>b</sup>, T. McCloskey<sup>b</sup>, and A.R. Chourasia<sup>c</sup><sup>a</sup>Physics Dept., Univ. of Missouri, Columbia, MO 65211<sup>b</sup>Materials Science Division, Argonne Natl. Lab., Argonne, IL<sup>c</sup>Dept. of Physics, Texas A&M Univ. -Commerce, Commerce, TX 75429

## ABSTRACT

Long wavelength spin waves in  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  CMR materials exhibit conventional Heisenberg dispersion ( $E = Dq^2 + \Delta$ ), but with an anomalous temperature dependence for the spin dispersion parameter  $D$ . For  $x \approx 0.33$ , near the optimum CMR composition,  $D$  remains approximately 50% of its  $T = 0\text{K}$  value before exhibiting a near-first-order transition at  $T_c$ , instead of following the expected power law decrease as  $T$  approaches  $T_c$ . No temperature hysteresis was observed in either  $D$  or in the magnetization. Compositions with  $x = 0.40$  and  $0.45$  show more conventional temperature renormalization.

Keywords: manganites, CMR, spin waves

## INTRODUCTION

Inelastic neutron scattering studies on polycrystalline samples of the perovskite-based colossal magnetoresistance manganites  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  with  $x$  near the optimal CMR composition  $x = 0.33$  show conventional spin wave dispersion ( $E = Dq^2 + \Delta$ ) at long wavelengths<sup>1,2</sup>.  $D$  is the spin wave stiffness constant and  $\Delta$  is a  $q = 0$  energy gap that may arise from anisotropy and is near 0 in these systems. The temperature dependence of  $D$  in an Heisenberg system is given by  $D(T) = D(T=0)[1 - (T/T_c)^{5/2}]$  at low  $T/T_c$ , with small higher order correction terms, and at temperatures approaching  $T_c$ ,  $D$  varies as  $[1 - (T/T_c)]^{\nu-\beta}$  where  $\nu - \beta \approx 0.34$ .

Previous results on polycrystalline  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  with  $x = 0.33$  by Lynn et al.<sup>1</sup> gave the unexpected result that  $D$  did not show the normal decrease on approach to  $T_c$ , rather it retained close to 50% of its  $T = 0\text{K}$  magnitude at  $T_c$ . Our results taken with finer temperature increments<sup>2</sup> show that  $D$  drops abruptly above  $0.98 T_c$  and exhibits a near-first-order transition at  $262\text{ K}$  that is also reflected in the magnetization determined with neutrons. No measurable hysteresis was found on warming and cooling in either quantity. Lynn et al.<sup>1</sup> also found that spin wave spectral weight was exchanged with an anomalous  $E = 0$  central mode, with an onset near  $60\% T_c$ , that increased rapidly in intensity at  $T_c$  and whose  $q$ -width corresponded to a  $13\text{ \AA}$  correlation range.

## SPIN WAVE BEHAVIOR

In an attempt to confirm if the anomalous  $T$ -dependence of  $D$  was present only near the optimal CMR compositions and thus might be associated with magnetic polaron formation<sup>1</sup>, we have investigated the spin waves in compositions ranging from  $x = 0.25$  to  $x = 0.45$  all in the ferromagnetic-metal range. Small  $q$  inelastic scattering techniques were employed on a triple axis spectrometer using an incident neutron energy of  $14.2\text{ meV}$ . Focusing effects present at small angle yielded an effective resolution for spin waves of about  $0.12\text{ meV}$ , which is approximately  $1/3$  of the elastic resolution. Useful spin wave data were obtained down to  $q = 0.04\text{ \AA}^{-1}$  with an upper  $q$  limit imposed by the spin wave

energies and kinematic considerations. These restrictions also precluded obtaining values of the spin wave stiffness parameter at low T.

Figure 1 illustrates the spin wave intensity versus energy transfer for the  $x = 0.36$  composition at several temperatures below the  $T_c = 269\text{K}$ . Above  $T_c$ , the energy-gain and energy-loss spin wave excitation peaks collapse into a single Lorentzian scattering peak at  $E = 0$ . Conventional Heisenberg dispersion was observed for the data of Figure 1, as well as that for all other compositions and temperatures.

No anomalous central peak phenomenon was found up to  $0.98 T_c$  for any composition except for  $x = 0.25$ . This compound exhibited a 4% difference in the peak temperature of the resistivity and the susceptibility peak, unlike others that show no measurable difference in these temperatures. One is tempted to associate this temperature difference and the occurrence of a central peak to a common physical origin.

The results for the temperature dependence of D for the 6 compositions derived from the dispersion curves are plotted in Figure 2. The solid lines are the result of a fit to above-mentioned power law at higher temperatures and to the  $T^{5/2}$  dependence at low T. It is noted that all compositions up through  $x = 0.36$  do exhibit the abrupt temperature renormalization of D near  $T_c$ , and consequently the fits produce unphysical values of the exponents  $\nu-\beta$ . The  $x = 0.40$  and  $x = 0.45$  compositions show a more normal T-dependence and yielded values of  $\nu-\beta = 0.36$  and  $0.38$  respectively. These higher compositions show a reduced CMR effect and are approaching the phase boundary at  $x = 0.5$  between the ferromagnetic metal and the antiferromagnetic insulator states. The  $T = 0\text{K}$  values of D given in the figure are extrapolated, due to the kinematic restrictions stated above.

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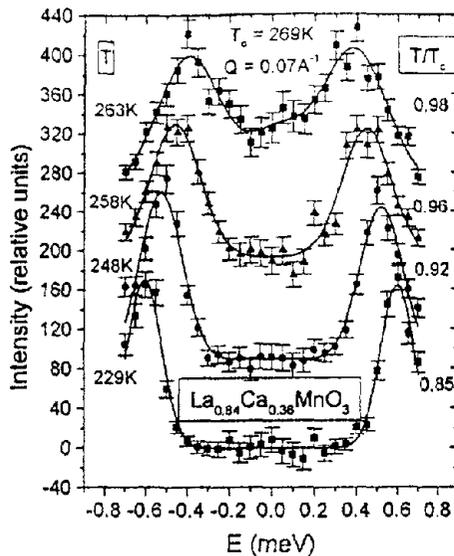


Figure 1. Spin wave intensity versus energy at a constant  $q = 0.07 \text{ \AA}^{-1}$  in  $\text{La}_{0.64}\text{Ca}_{0.36}\text{MnO}_3$  for several temperatures below  $T_c$ . No evidence of a central mode anomaly is seen below  $0.98 T_c$ , and the spin wave integrated intensity rises with temperature in accordance with Boltzman statistics.

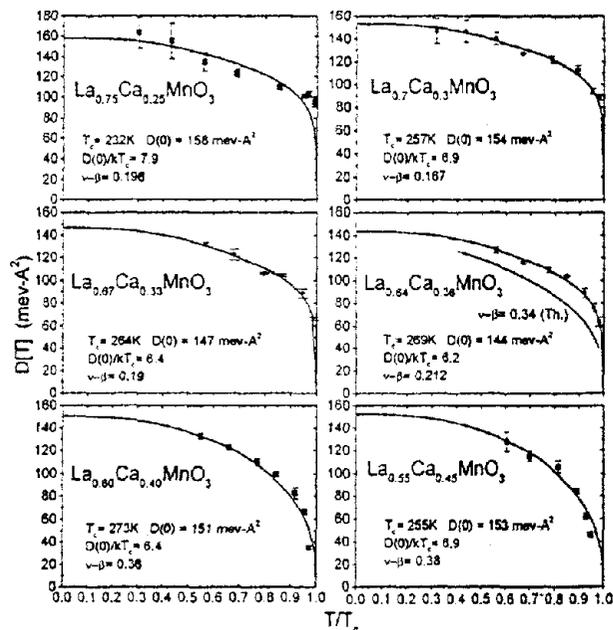


Figure 2. Temperature dependence of the spin wave stiffness parameter  $D$  in  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  for six compositions in the ferromagnetic metal regime. The solid lines are the result of a fit to a power law at high  $T$  and to a  $T^{5/2}$  dependence at low  $T$  (see text). The curve for the theoretically expected power law exponents  $\nu-\beta = 0.34$  is shown for comparison in the  $x = 0.36$  frame.

## FIGURE CAPTIONS

Figure 1. Spin wave intensity versus energy at a constant  $q = 0.07 \text{ \AA}^{-1}$  in  $\text{La}_{0.64}\text{Ca}_{0.36}\text{MnO}_3$  for several temperatures below  $T_c$ . No evidence of a central mode anomaly is seen below  $0.98 T_c$ , and the spin wave integrated intensity rises with temperature in accordance with Boltzman statistics.

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