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Low-temperature specific heat of $La_{0.78}Pb_{0.22}MnO_{3-\delta}$ manganite

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Abstract

The specific heat of the perovskite manganite $La_{0.78}Pb_{0.22}MnO_{3-\delta}$ was measured from 0.5 to 25 K. The results yield values of the electronic density of states, the Debye temperature, and the spin-wave stiffness constant of the compound. The magnetic contribution was found to be consistent with the existence of a gap in the spin-wave spectrum, and a phonon term in T^5 was needed to fit the data.

Keywords: Manganite; Colossal magnetoresistance; Specific heat; Spin waves

In recent years, a lot of attention has been focused on perovskite-type manganite systems, due to the discovery of colossal magnetoresistance (CMR) effects, and a great variety of magnetic and transport properties observed in these compounds [1]. Among the various doped lanthanum-manganese oxides that exhibit CMR properties, $La_{1-x}Pb_xMnO_3$ has been relatively less investigated. In the present study, we have used specific-heat measurements to probe the low-temperature excitations in $La_{0.78}Pb_{0.22}MnO_{3-\delta}$, a metallic ferromagnet with $T_c = 345$ K. The simplicity of the low-temperature spin waves [2] in this undistorted perovskite sample makes it a representative candidate for the investigation of some fundamental properties in double-exchange ferromagnets.

The measured samples are large single crystals, obtained by the high-temperature solution growth method. The chemical composition was verified with an ED spectrometer, and found to be $La_{0.775(5)}Pb_{0.224(9)}MnO_{2.713(20)}$. X-ray analysis confirmed a cubic perovskite structure with cell parameters $a = b = c = 3.894$ Å. Magnetization, resistivity and magnetoresistance were measured with a quantum design PPMS system. Differential scanning calorimetry was performed on a TA instruments equipment. The specific-heat results were obtained from 0.5 to 25 K, with an automated quasi-adiabatic pulse technique, in a home-made ^{3}He calorimeter.

The behavior of the para-ferromagnetic and metal–insulator transition was characterized with magnetic, transport, and thermal data. Fig. 1a shows the zero-field cooled magnetization (M) as a function of temperature, where a sharp ferromagnetic transition can be observed. The inset of Fig. 1a shows isothermal M vs. H data measured at 10 K. The saturation value of 3.75 μ_B virtually coincides with the value expected from the spin contribution arising from Mn^{3+} and Mn^{4+} ions (3.78 μ_B). Resistivity measurements at various applied fields, plotted in Fig. 1b, show that a metal–insulator transition occurs simultaneously with the ferromagnetic order. Magnetoresistance values higher than 50% are obtained with an applied field of 9 T, as shown in the inset of Fig. 1b. High-temperature specific-heat data, plotted in the inset of Fig. 2, also reveal a distinct anomaly at 346 K, coinciding with the magnetic and transport transition.

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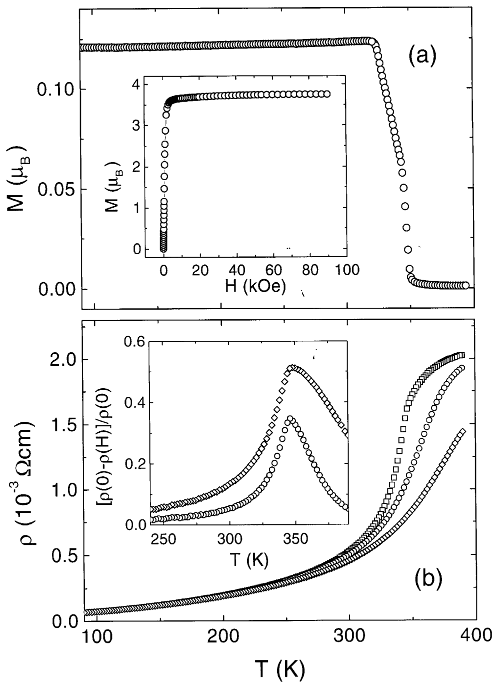

Fig. 1. (a) Zero field cooled magnetization (M) of $La_{0.78}Pb_{0.22}MnO_{3-\delta}$, measured with $H = 50\,Oe$. The inset shows data of M vs. H measured at 10 K. (b) Resistivity (ρ) vs. temperature for $H = 0$ (squares), 3 (circles) and 9 T (diamonds). The inset shows the magnetoresistance ratio, defined as $[\rho(0) - \rho(H)]/\rho(0)$, at $H = 3$ and 9 T.

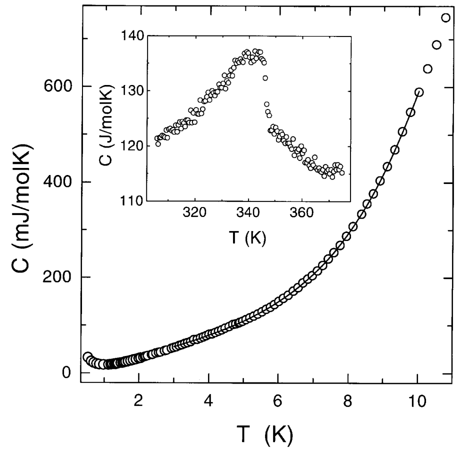

Fig. 2. Low-temperature specific heat of $La_{0.78}Pb_{0.22}MnO_{3-\delta}$. The solid line is a fit to the data as discussed in the text. The inset shows results of high-temperature specific heat in the same compound.

The low-temperature specific-heat data and the fitted curve are shown in Fig. 2. The results were fitted considering a linear term γT arising from free electrons, a $\beta T^3 + \eta T^5$ contribution from lattice vibrations, and a magnetic part. The latter was expressed as $C_{mag} = \delta T^{3/2}(1 + 4\Delta/5T + 4\Delta^2/15T^2)\exp(-\Delta/k_B T)$. The term $\delta T^{3/2}$ is commonly used to describe the contribution from ferromagnetic spin-wave excitations. The exponential factor and the other terms appear when one considers a magnon dispersion relation of the form $\omega(k) = \Delta + Dk^2$. This is consistent with previous neutron-scattering experiments on $La_{0.7}Pb_{0.3}MnO_3$ [2], which detected an energy gap $\Delta/k_B = 25\,K$. At lower temperatures, the specific heat data shows an upturn due to a Schotky contribution. Our results were fitted in the region from 3 to 10 K. Keep-ing Δ/k_B fixed to the value of 25 K, we obtained $\gamma = 15\,mJ/molK^2$, $\beta = 0.23\,mJ/molK^4$, $\eta = 0.0021\,mJ/molK^6$ 0.41 mJ/molK$^{5/2}$. The linear term gives a density of states of $3.8 \times 10^{24}\,eV/mol$ at the Fermi level. This is higher than that observed in other manganite samples, although a large linear term associated with charge localization has been previously reported [1,3]. The cubic term gives a Debye temperature $\theta_D = 348\,K$, similar to various perovskite systems. The magnetic coefficient δ is related to the spin-wave stiffness constant D, yielding a value of $186\,meV\,\mathring{A}^2$. From the usual relation $D = 2SJa^2$, where S is the spin and a is the lattice parameter, one can extract an effective exchange interaction J compatible with the observed critical temperature of this compound.

In summary, we have measured the low-temperature specific heat of $La_{0.78}Pb_{0.22}MnO_{3-\delta}$, a typical double-exchange ferromagnet. The results allowed us to estimate the electronic density of states at the Fermi level, the Debye temperature, and the spin-wave stiffness constant for this compound. The magnetic contribution can be described by ferromagnetic spin-wave excitations with a gap. It is worth noting that the almost constant spontaneous magnetization in the low-temperature region and its sharp drop near T_c (Fig. 1a) are also indicative of magnetic anisotropy, in agreement with the presence of a gap in the spin-wave spectrum [4].

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