Low-temperature field-dependent magnetization of $La_{0.7}Sr_{0.3}MnO_3$

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We report measurements of the magnetization of single crystalline $La_{0.7}Sr_{0.3}MnO_3$ for temperatures $T<25$ K and in fields of 1 and 3 T. This material is a ferromagnetic metallic oxide which exhibits colossal magnetoresistance. We find that the field-dependent magnetization decreases with temperature in a manner consistent with spin-wave excitations. That is, in zero field the magnetization would vary as $M(0)-M(T)\propto T^{3/2}$, as opposed to T^2 which might be expected with single-particle excitations. From this we are able to extract a spin-wave stiffness value of 154 ± 5 meV Å^2 which is in excellent agreement with recent neutron scattering and spin-wave resonance results. [S0163-1829(97)03309-2]

Recently, there has been a great deal of interest in materials showing colossal magnetoresistance (CMR). This interest has been both experimental[1] and theoretical.[2] One prominent class of CMR materials is the family $La_{1-x}A_xMnO_3$, where the divalent A is Ba, Ca, or Sr, and $0\leqslant x\leqslant 0.5$. The largest CMR effects occur near $x\sim\frac{1}{3}$. At low temperatures these materials are ferromagnetic, although the undoped material ($x=0$) can become antiferromagnetic with slight changes of stoichiometry.[3] At the magnetic critical temperature T_c, they become paramagnetic. Doped members of the family ($x\neq 0$ or 1) are metallic below T_c, but experience changes in conductivity at temperatures $T\sim T_c$. When A is Ba or Ca, the conductivity above T_c is characteristic of an insulator or semiconductor, whereas the Sr-doped material remains metallic. The fundamental origin of the CMR effect and the changes in conductivity near T_c are not well understood at present.

Traditionally, the electronic and magnetic properties of these manganites were explained by the double-exchange model,[4] where electrons transfer between Mn^{3+} and Mn^{4+} ions on adjacent sites. However, a recent calculation[5] showed that the resistivity change in an applied magnetic field could not be explained by double exchange alone, and the authors proposed that a Jahn-Teller-type electron-phonon coupling must also play an important role. Moreover, there are some materials which evidently show the CMR effect without either the double-exchange or Jahn-Teller mechanisms.[6]

To better understand the magnetic and electronic properties of these materials, it is desirable to measure the fundamental properties of the ferromagnetic state. In this paper we report measurements of the low-temperature magnetization of a single $La_{0.7}Sr_{0.3}MnO_3$ crystal with $T_c=360$ K. We find that the magnetization is consistent, when the proper field dependence is taken into account, with the excitation of spin waves of magnetic stiffness $D=154\pm5$ meV Å^2. The stiffness is defined by the low-momentum spin-wave dispersion relation

$$\epsilon(q)=\Delta+Dq^2 \quad (q\rightarrow 0), \tag{1}$$

where ϵ is the spin-wave energy, q is the momentum wave vector, and Δ is a gap energy arising from anisotropy or applied magnetic field H. This value of D is fully consistent with recent neutron-scattering[7] and spin-wave resonance[8] measurements.

These results imply that these materials should follow the ''Bloch $T^{3/2}$ law,'' i.e., that the zero-field magnetization $M(T,H=0)$ should have a temperature dependence of

$$M_0-M(T,0)=(\text{const})\times T^{3/2}, \tag{2}$$

where $M_0=M(0,0)$ is the full saturated magnetization. This contrasts with the findings of Snyder *et al.*[9] which indicate a T^2 temperature dependence, characteristic of individual particle (Stoner-type) excitations.[10]

In our measurements we restricted temperatures to $T<25$ K so that $T/T_c<0.1$ throughout the range of analysis, and measured in two values of applied field, 1 and 3 T. Generally speaking, the Bloch law is expected to hold out to $T/T_c\sim 0.2$ in systems where it is applicable.

Our 41.2-mg sample was cut from a larger single crystal provided by the Moscow State Steel and Alloys Institute. The crystal itself was grown using the floating zone method, and energy-dispersive x-ray analysis (EDAX) showed it to be highly pure and verified the stoichiometry. X-ray diffraction indicated a high degree of crystalline quality, and a demonstration of magnetic quality came from the narrowness of a ferromagnetic resonance with a linewidth of only 5 mT.[11]

Measurements were made in a Quantum Design superconducting quantum interference device (SQUID) magnetometer where the sample was cooled to 5 K or below in zero field. Then the working field was applied, and the magnetization was measured with the sample warming. This method is more sensitive for measuring temperature variations than the standard method for testing the Bloch law. The Bloch law, it must be remembered, is valid only for $H=0$. In the

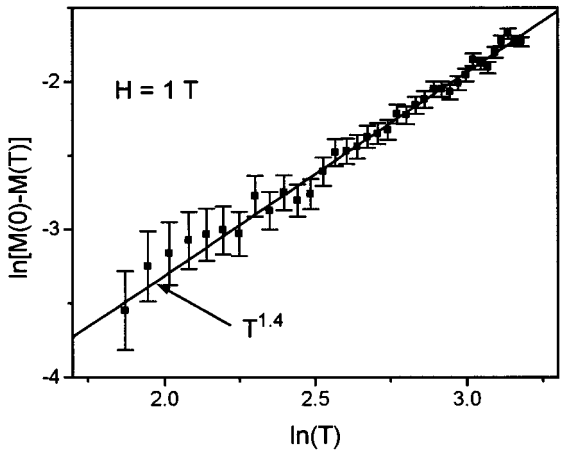

FIG. 1. Magnetization M vs temperature T for $H=1$ T. The line is a weighted best fit to Eq. (3) of the text. The 3-T data are similar, but a best fit indicates $T^{1.7}$.

standard method,[12] M is measured as a function of H at fixed T. Then, the data are extrapolated back to $H=0$ to construct $M(T,0)$. Unfortunately, although the SQUID is highly sensitive, the uncertainty in $M(T,0)$ produced by this extrapolation in H dwarfs the $\sim$0.2% signal variation with temperature in the range 5–25 K.

At each field, 4–6 runs were taken without removing the sample from the magnetometer. Typically, each run would be displaced from the previous one by a random amount within $\sim$0.08% of $M(0,H)$. Within the scatter of the data, however, all runs were parallel. Removing the sample from the magnetometer could produce a change in the measured magnetization of up to a few percent, owing to slight changes in sample orientation. In each case, the three lowest temperature points were discarded to negate any effects of system startup transients, and the individual runs were averaged to form the final data set. Finally, a small background signal, measured at both fields with no sample in the magnetometer, was subtracted off.

For each field a second order polynomial was used to extrapolate $M(T,H)$ back to $T=0$. Care is required during this step since variations in $M(0,H)$ can affect the results. Using this method, $M(0,H)$ was determined to within 0.006%, and values of $M(0,H)$ agreed with a fully aligned species consisting of 0.7 Mn^{3+} ions (spin 2) and 0.3 Mn^{4+} ions (spin $\frac{3}{2}$) per unit cell. Given the dimensions of our particular sample, the saturation field was 0.55 T.

If we assume that the low-temperature magnetization follows the form of

$$M(0,H)-M(T,H)=(\text{const})\times T^{\alpha}, \tag{3}$$

where α is to be measured, then the result is similar to that depicted in Fig. 1. Figure 1 shows the magnetization for $H=1$ T on a logarithmic scale. A weighted fit to a simple power law is seen to be a reasonable characterization of the data, and produces a value of $\alpha=1.4\pm0.1$. At first glance, this appears to be a confirmation of the Bloch law.

However, we have yet to properly take account of the presence of the field. Indeed, if a similar analysis is performed on the 3-T data, then the fit is equally good, but α turns out to be 1.7 ± 0.1. The key is to understand the effect of applied field on the spin-wave dispersion relation in Eq. (1). In Eq. (1) the gap energy Δ is given by

$$\Delta=\Delta_0+g\mu_B(H-NM), \tag{4}$$

where Δ_0 is the intrinsic gap, $g=2$ is the Landé g factor, μ_B is the Bohr magneton, N is the demagnetization factor, and M is the measured magnetization of the sample. In our case we can simply take M as M_0, as the sample was essentially fully magnetized at all times. The derivation of the Bloch law assumes that $\Delta=0$. Indeed, neutron scattering has placed an experimental upper limit on Δ_0 of 40 μeV for other members of the $La_{1-x}A_xMnO_3$ family,[7] and a direct measurement of the anisotropy field in our sample found it to be less than 0.02 T, or $\Delta_0<2.5$ μeV.[13] So the assumption that $\Delta_0=0$ is valid, but the application of H forces a nonzero Δ, in which case the Bloch law must be modified. For temperatures below $\sim0.2T_c$, the momentum of thermally excited spin-waves is low enough that Eq. (1) is a good approximation to the dispersion relation. Using the standard spin-wave picture, we find that in this limit the magnetization becomes

$$M(0,H)-M(T,H)=g\mu_B\left(\frac{k_BT}{4\pi D}\right)^{3/2}f_{3/2}(\Delta/k_BT), \tag{5}$$

where k_B is Boltzmann's constant, and $f_{3/2}(y)$ is given by

$$f_P(y)=\sum_{n=1}^{\infty}\frac{e^{-ny}}{n^P}. \tag{6}$$

Equation (5) reduces to the Bloch law when H and Δ are zero.

Our sample is fairly flat, being a chip of dimensions $\sim3\times3\times0.5$ mm^3, and from the saturation field we estimated the demagnetizing factor to be $N\sim80\%(4\pi)$. With a magnetization of 95 emu/g, assuming a lattice spacing of 3.92 Å gives a total demagnetizing field NM of about 0.6 T. Thus, Δ/k_BT ranges from 0.02–0.7 in our experiment, and is not necessarily small.

Over the restricted range of temperature in our experiment, Eq. (5) is well approximated by Eq. (3) with some effective exponent α_{eff}. Figure 2 shows the expected form of α_{eff} as a function of applied field, compared with the two values measured from the data. As can be seen, at high fields α_{eff} takes on the value 2. This could be one reason why high-field magnetization measurements on $SrRuO_3$ show a T^2 dependence.[14]

The existence of spin-waves in the $La_{1-x}A_xMnO_3$ family is not in doubt. They have been unambiguously detected in inelastic neutron scattering, both at low momentum[7] and throughout the Brillouin zone,[15] and in spin-wave resonance[8] experiments. However, the existence of well-defined spin waves in metallic ferromagnets does not insure that the Bloch $T^{3/2}$ law is followed,[10] particularly in cases where ferromagnetism is weak.[16] Weak ferromagnets are characterized by a low ratio of saturated moment p_s to the total effective moment p_{eff} of the system. The latter is determined from the Curie constant,[12] and weak ferromagnets typically have $p_s/p_{\text{eff}}<0.2$. It should be noted that both $La_{1-x}A_xMnO_3$ ($p_s/p_{\text{eff}}=1$) and $SrRuO_3$ [$p_s/p_{\text{eff}}=0.87$ (Ref. 14)] are strong ferromagnets.

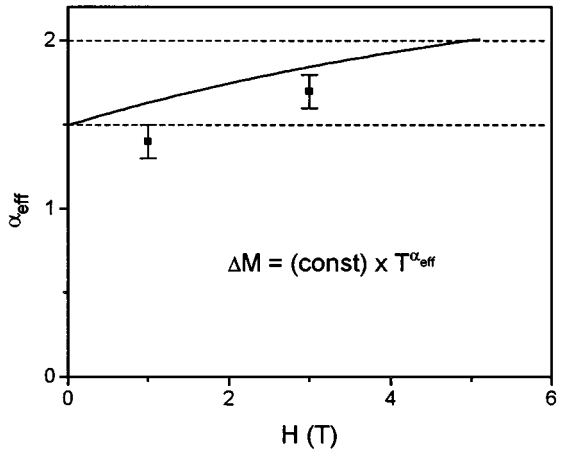

FIG. 2. The solid line indicates the effective exponent α_{eff} vs field H computed by approximating Eq. (5) by Eq. (3) over the temperature range 5–25 K. The two individual points are from the data, and the dashed lines indicate the values $\frac{3}{2}$ and 2.

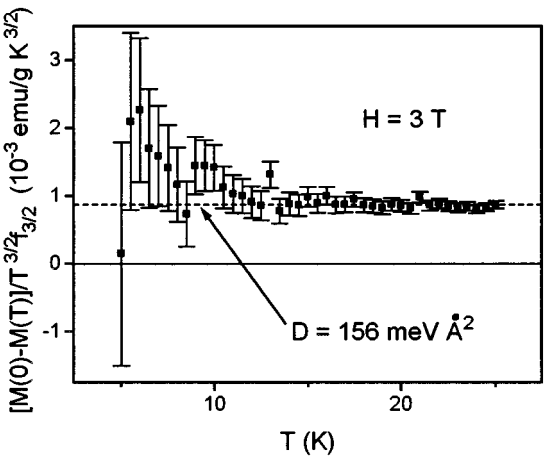

FIG. 3. 3-T data plotted in such a way as to be a constant according to Eq. (5) of the text. The 1-T data are similar. The weighted-fit value of the constant gives a magnetic stiffness $D=156$ meV Å^2, which is within 3% of the 1-T value and in excellent agreement with neutron-scattering and spin-wave resonance data.

If the field dependence of Eq. (5) is divided out of the data, then a best fit for both the 1 and 3 T data throughout the entire temperature range yields a remnant temperature dependence of $\sim T^{1.2}$. However, owing to the large relative error of the lowest temperature points, this is not inconsistent with $T^{3/2}$. For example, if only the data above 13 K are considered, the remnant temperature dependence for 3 T becomes $T^{1.35\pm0.13}$. Figure 3 shows the 3-T data plotted in the form of $[M(0,H)-M(T,H)]/[T^{3/2}f_{3/2}(\Delta/k_BT)]$ vs T for the whole temperature range. According to Eq. (5), this should be a constant, and the weighted best-fit to Eq. (5) is shown for comparison. The resulting value for D is 156 meV Å^2, and is within 3% of the 1-T value of 151 meV Å^2. Averaging these values, and accounting for the uncertainty in determining $M(T=0,H)$, gives $D=154\pm5$ meV Å^2 which is in excellent agreement with the neutron-scattering[7] and spin-wave resonance[8] results.

At low temperatures, the magnetic contribution to the sample heat capacity should also vary as $(T/D)^{3/2}$ times a function of Δ/k_BT.[17] The values of D are such that it would be very difficult to resolve the magnetic contribution in specific heat data. Indeed, this was found to be the case for barium- and calcium-doped manganite samples,[18] as well as the $La_{0.7}Sr_{0.3}MnO_3$ crystal in a separate experiment.

In conclusion, we have measured the field-dependent magnetization of a single crystal of the CMR material $La_{0.7}Sr_{0.3}MnO_3$ at temperatures below $0.1T_c$. We have found that the magnetization is characteristic of a simple spin-wave excitation spectrum, with no evidence of single-particle Stoner-type excitations. The measured value of magnetic stiffness, $D=154\pm5$ meV Å^2, is in excellent agreement with values previously measured by other methods.

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