Magnetic phase transition in $La_{0.7}Sr_{0.3}MnO_3$: Microwave absorption studies

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Using a special technique for measuring the saturation magnetization at temperatures close to the Curie point, we demonstrate that in $La_{0.7}Sr_{0.3}MnO_3$ the magnetic phase transition is continuous. [S0163-1829(97)02805-1]

$La_{0.7}Sr_{0.3}MnO_3$, a prototypical colossal magnetoresistance perovskite, has generated much interest in recent work. It has a ferromagnetic transition near 360 K. High-quality samples have the lowest resistivity among all the manganites and essentially metallic resistivity in both the paramagnetic and ferromagnetic phases.[1] Thus it is the most likely candidate for a simple double-exchange system.[2] As regards its magnetic properties, the low-lying excited states are spin waves as shown by neutron scattering[3] and spin-wave resonance.[4] However, the nature of the magnetic transition in these systems is far from settled. Although some of the results[3,5] point to second-order behavior, there have also been suggestions[6,7] that the magnetic transition is first order in that there is a discontinuity in the magnetization as well as a thermal hysteresis. In order to help resolve this question, one needs careful measurements of the spontaneous magnetization M_S in the neighborhood of the Curie temperature T_C. A single-crystal sample is required because in many perovskite specimens it has been found[8] that the material consists of several magnetic subnetworks with a sizable spread of T_C values. This report presents methods for measuring the magnetization of a single crystal of $La_{0.7}Sr_{0.3}MnO_3$ covering the temperature range 310–360 K; i.e., in the neighborhood of the magnetic phase transition. It turns out that the temperature dependence of M_S is well described by $(T_C-T)^\beta$ with $T_C = 360\pm 1$ K and $\beta = 0.45\pm 0.05$.

A single crystal of nominally $La_{0.7}Sr_{0.3}MnO_3$ was made by the floating-zone technique at the Moscow State Steel and Alloys Institute. The samples used here were cut from the boule to yield parallelpipeds of dimensions roughly $3\times 5 \times 0.5$ mm^3. The flat surfaces were polished using 1-μm diamond paste. Both electron dispersive x-ray analysis and x-ray diffraction showed the crystal to be homogeneous.

The low-field (0.3 mT) ac susceptibility (χ_{ac}) transition (Fig. 1) is quite sharp, extending from 352 to $\sim$ 360 K. Note that there is no thermal hysteresis.

The ferromagnetic resonance linewidth Γ (Refs. 9 and 10) is small ($\sim$ 50 Oe at 340 K and 10 GHz), and its temperature dependence near 360 K reveals that any distribution in T_C values is minimal.[8] Although Γ increases slightly on lowering of temperature, an indication of a small magnetic inhomogeneity, the magnetic integrity of the crystal is sufficiently good to ensure that the results described below represent intrinsic behavior.

The magnetization was investigated by three techniques employing microwave absorption measurements at several frequencies. The first method[11] consists of placing a small paramagnetic (PM) marker [a grain of diphenylipicrylhydrazyl (DPPH)] at the center of the surface of the crystal and measuring its (PM's) electron spin resonance (ESR) field at 9.18 GHz. A shift in the measured resonance position arises from the local demagnetizing field, of the sample, which is directly proportional to the saturation magnetization. Further, since the crystal shape is well defined, the demagnetizing factor D at the marker site is 0.056. Thus one can compute M_S, and the present results are in good agreement with those reported in Refs. 2 and 3.

The second and third techniques are based upon a recently proposed idea[10] suggesting the use of the ferromagnetic antiresonance (FMAR) phenomenon, the minimum which should occur in the field-induced mircowave absorption in a

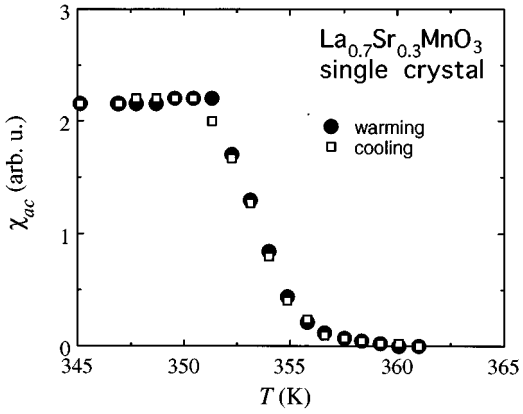

FIG. 1. ac susceptibility of a $La_{0.7}Sr_{0.3}MnO_3$ single crystal as a function of temperature. Note the sharp rise consequent on the onset of ferromagnetism. The field amplitude $\mu_0 H$ was 0.3 mT at a frequency of 140 Hz. The cooling and the warming cycles agree quite closely with each other.

bulk metallic ferromagnetic specimen. For a parallelepiped, the FMAR results when

$$\omega/\gamma=\mu_0(H'+M_S), \tag{1}$$

where ω is the angular frequency, γ the gyromagnetic ratio, and H' the internal field. Near T_C, M_S varies rapidly with temperature T. Hence, by studying the microwave absorption at a constant field as a function of T, one expects to observe a minimum at a characteristic temperature where the FMAR condition [Eq. (1)] is satisfied.

The FMAR effect can be modeled using the Landau-Lifschitz-Gilbert equation,[10] from which the dynamic susceptibility χ for a monodomain ferromagnetic ''disk'' can be written as

$$\chi=\frac{\mu_0 M_S[\mu_0 H'+\mu_0 M_S+i\Gamma]}{(\mu_0 H'+i\Gamma)[\mu_0 M_S+i\Gamma]-\left(\frac{\omega}{\gamma}\right)^2}. \tag{2}$$

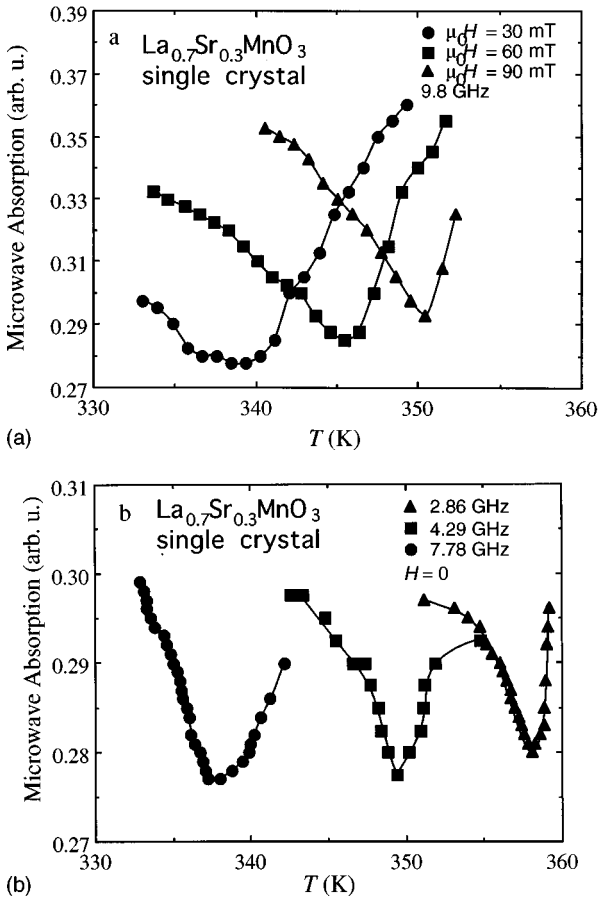

FIG. 2. (a) Temperature dependence of the microwave absorption of a $La_{0.7}Sr_{0.3}MnO_3$ single crystal at 9.8 GHz in the vicinity of the magnetic transition. Applied field values were as indicated. The lines serve as guides to the eye. The minima which satisfy the FMAR condition [Eq. (1)] are defined to about 1 K. For presentation purposes, the curves have been shifted vertically by arbitrary amounts, and the absolute value has no significance. (b) Temperature dependence of the zero-field microwave absorption of a $La_{0.7}Sr_{0.3}MnO_{3f}$ single crystal at 2.86, 4.29, and 7.78 GHz and T close to T_C. The lines serve as guides to the eye.

The microwave losses are proportional to $\sqrt{-i\mu}$ where μ is the dynamic permeability $(=1+\chi)$. Calculations using Eq. (1) show that in order to see the T-dependent dip in absorption at a constant applied field, Γ must be less than $0.05\omega/\gamma$. For larger linewidths, the FMAR effect, in this representation, is not very strong. In fact, for $\Gamma\sim 0.2\omega/\gamma$, as in the case of bulk ceramic $Nd_{0.7}Sr_{0.3}MnO_3$,[8] one can expect only a 5% decrease in the absorption. Therefore, it is understandable that the 10 and 60 GHz absorption data for the $Nd_{0.7}Sr_{0.3}MnO_3$ agreed[12] even though the FMAR condition was satisfied at the 10 GHz but not at 60 GHz.

By locating the minimum in the T dependence of the microwave absorption, M_S was obtained at $\mu_0 H$ ranging from 30 to 270 mT at 9.8 GHz [Fig. 2(a)] and at $H=0$ for frequencies ranging from 2 to 11 GHz [Fig. 2(b)]. The minima are definable to about 1 K. Note that by either lowering the frequency or increasing the applied field, the dip in the absorption is sharpened, reflecting the fact that M_S varies more rapidly as T approaches the Curie point.

Strictly speaking, Eq. (2) can only be used in the single-domain state, and the interpretation of the zero-field results is not so straightforward. However, we have used Eq. (2) and a simple convolution to make calculations for several different domain configurations, and it turns out that, in every case, the drop in the zero-field absorption occurs when $\omega/\gamma=\mu_0 M_S$. For the present samples and temperatures of interest, the difference between the internal field H' and the applied field H is negligible in comparison to M_S, and we are confident that simple use of the nonzero H case FMAR condition $[\omega/\gamma=\mu_0(H+M_S)]$ applies.

Shown in Fig. 3 are the resulting $\mu_0 M_S$ values as a function of T. The solid curve is a least-squares fit to the relation $M_S\propto(T_C-T)^\beta$. It yields a critical exponent β of 0.45 ± 0.05 with $T_C=361\pm1$ K. This value of β is in agreement with the value (0.495) given by Morrish[5] for a $La_{0.56}Pb_{0.44}MnO_3$ crystal, but is rather larger than the value[3] found for a $La_{0.7}Sr_{0.3}MnO_3$ crystal by neutron scattering ($\beta=0.295$). Regardless, if this were indeed a first-order transition, such a fit

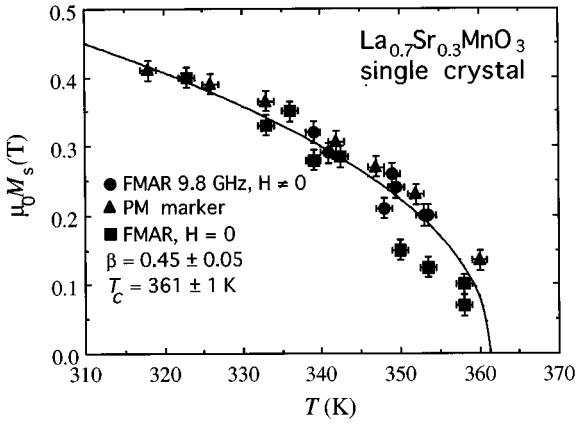

FIG. 3. Magnetization of a $La_{0.7}Sr_{0.3}MnO_3$ single crystal obtained from the three methods described in the text: triangles, shifted ESR of PM marker; circles, minimum in the T-dependent 9.8-GHz absorption at finite field [Fig. 2(a)]; and squares, minimum in the T-dependent absorption at $H=0$ and different microwave frequencies [Fig. 2(b)]. The full curve represents a fit to $M_S\propto(T_C-T)^\beta$ with the fit parameters given in the figure.

would not work as the M vs T curve would have a step at T_C. Also, we found that for any particular measurement, there is no discernible difference between data taken while cooling or warming. Both the fit to a scaling function and the lack of thermal hysteresis are good evidence that the magnetic transition is continuous.

In conclusion, we have measured the magnetization near the Curie point of $La_{0.7}Sr_{0.3}MnO_3$ using microwave absorption methods. We have found M_S varying as $(T_C-T)^{\beta}$ and no thermal hysteresis, suggesting that the magnetic transition is second order with essentially a mean-field exponent. We have demonstrated that, in the neighborhood of T_C, the technique of following the T dependence of the microwave absorption (FMAR) at various frequencies in a bulk metallic ferromagnet is an effective tool for measuring the spontaneous magnetization with the application of little or no field. It is useful to note the similarity of the present method to the study of nuclear magnetic resonance[13] as well as the caliper resonance[14] in a second-sound cavity in the neighborhood of their respective second-order transitions.

Single crystals were grown by Y. M. Mukovskii and S. G. Karabashev at the Moscow State Steel and Alloys Institute and by A. M. Balbashov at the Moscow Power Engineering Institute.

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