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Determination of the magnetization scaling exponent for single-crystal $La_{0.8}Sr_{0.2}MnO_3$ by broadband microwave surface impedance measurements

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Employing a broadband microwave reflection configuration, we have measured the complex surface impedance, $Z_S(\omega,T)$, of single-crystal $La_{0.8}Sr_{0.2}MnO_3$, as a function of frequency (0.045–45 GHz) and temperature (250–325 K). Through the dependence of the microwave surface impedance on the magnetic permeability, $\hat{\mu}(\omega,T)$, we have studied the local magnetic behavior of this material, and have extracted the spontaneous magnetization, $M_0(T)$, in *zero applied field*. The broadband nature of these measurements and the fact that no external field is applied to the material provide a unique opportunity to analyze the critical behavior of the spontaneous magnetization at temperatures very close to the ferromagnetic phase transition. We find a Curie temperature $T_C = 305.5 \pm 0.5$ K and scaling exponent $\beta = 0.45 \pm 0.05$, in agreement with the prediction of mean-field theory. We also discuss other recent determinations of the magnetization critical exponent in this and similar materials and show why our results are more definitive.

Since the recent discovery of large negative magnetoresistance in the manganite perovskites $La_{1-x}A_xMnO_3$ (where A is typically Ca, Sr, or Ba),[1,2] much attention has been paid to understanding the properties of these materials.[3] In addition to being potentially useful in technological applications, these so-called colossal magnetoresistive (CMR) oxides provide a system in which to study electronic and magnetic correlations and the interplay between magnetism and transport properties. In particular, a series of measurements in recent years have focused on the critical behavior of the magnetization in the vicinity of the ferromagnetic phase transition.[4–11] These experiments, employing a variety of techniques, have given widely varying values of the magnetization scaling exponent β ranging from about 0.3 to 0.5. This range encompasses both the long-range interactions of mean-field theory ($\beta = 0.5$),[12,13] and the values of β which result from calculations based on shorter range interactions, such as the Ising and Heisenberg models ($\beta = 0.325$ and 0.365, respectively).[14]

In this paper we present the results of broadband, nonresonant microwave surface impedance measurements, in which we have quantitatively determined the complex surface impedance, $\hat{Z}_S = R_S + iX_S$, of $La_{0.8}Sr_{0.2}MnO_3$ single crystals over three decades in frequency, and as a function of temperature. In contrast to conventional magnetization measurements, which typically require the application of an external magnetic field, this technique allows us to extract the temperature dependence of the static spontaneous magnetization in *zero applied field*. We have found that the spontaneous magnetization is zero above T_C and rises continuously below T_C in a manner which is well described by the theory of critical phenomena near a second-order phase transition. From these data we are able to determine the scaling exponent β, and find that it is consistent with the value predicted by mean-field theory.

The single crystals of $La_{0.8}Sr_{0.2}MnO_3$ used in this study were grown by the floating-zone technique[15] and the stoichiometry and structural integrity have been checked by x-ray diffraction and energy dispersive x-ray analysis. Disk-shaped samples were cut from a 4 mm diameter rod, and resistivity, ac susceptibility, and dc magnetization measurements have been reported earlier on samples cut from the same boule.[7] $La_{0.8}Sr_{0.2}MnO_3$ has a ferromagnetic phase transition with a Curie temperature T_C of approximately 305 K. It is well established that the low-temperature phase is a ferromagnetic metal, while above T_C the system is paramagnetic, and the resistivity exhibits a negative slope with respect to temperature. The resistivity has a maximum around $T_p = 318$ K, significantly above T_C, which is typical in these manganite materials.[7]

In order to determine the temperature and frequency dependence of the complex surface impedance we have measured the complex reflection coefficient. We have reported the details of the experimental geometry elsewhere,[16] and will therefore give only a brief overview of the technique here. A phase-locked signal from an HP8510C vector network analyzer (45 MHz–50 GHz) is sent into a coaxial transmission line which is terminated by the sample inside a continuous flow cryostat. The amplitude and phase of the reflected signal are measured as functions of frequency and temperature, and the complex reflection coefficient, $\hat{S}_{11}(\omega,T)$, is determined as the ratio of the reflected to incident signals. The complex surface impedance of the terminating material can be calculated from the reflection coefficient as follows: $\hat{Z}_S/Z_0 = (1+\hat{S}_{11})/(1-\hat{S}_{11})$, where $Z_0 = 377\ \Omega$ is the impedance of free space. Due to the phase-sensitive detection capabilities of the network analyzer, it is possible to extract *both* the surface resistance $R_S(\omega)$ and the surface reactance $X_S(\omega)$, and the well-defined geometry allows for *quantitative* evaluation of these material parameters, opening a unique window to dynamical processes within the material.

In the presence of a magnetic field the microwave properties of ferromagnetic materials are characterized by two

distinct features which result from the dispersion of the complex magnetic permeability $\hat{\mu}(\omega)=\mu_1(\omega)-i\mu_2(\omega)$: ferromagnetic resonance (FMR) and ferromagnetic antiresonance (FMAR).[17] At the FMR frequency, ω_r, the surface resistance, $R_S(\omega_r)$, shows a maximum due to a maximum in the imaginary part of the permeability. At the same frequency, the real part of the permeability has a zero crossing with negative slope, as does the surface reactance, $X_S(\omega_r)$. In order to satisfy the condition $\mu_1(\omega\rightarrow\infty)=1$, it is necessary that there be another zero crossing, with positive slope, at a frequency $\omega_{ar}>\omega_r$. For a ferromagnetic metal, this zero in μ_1 leads to a *reduction* in R_S below the value it would have for a nonmagnetic metal with the same resistivity. This suppression of R_S in the vicinity of ω_{ar} is commonly referred to as the ferromagnetic *anti*resonance. Both ω_r and ω_{ar} depend not only on the externally applied field but also on the local internal magnetization of the material, and measurements of the microwave surface impedance therefore yield information about the magnetization of the material under study.

The precise dependence of ω_r and ω_{ar} on the magnetization M_0 can be determined by starting from the Landau-Lifshitz-Gilbert equation of motion for the magnetization vector in the presence of both a static magnetic field H_0 and an oscillatory microwave field.[17–19] The complex dynamic susceptibility of such a system can be written in the following form:[17]

$$\hat{\chi}(\omega)=\frac{\hat{\mu}(\omega)}{\mu_0}-1=\frac{\omega_M[(\omega_0+i\Gamma)+\omega_M]}{\omega_r^2-\omega^2+i\Gamma[2\omega_0+\omega_M]}, \qquad (1)$$

where $\omega_M=\gamma\mu_0 M_0, \omega_0=\gamma\mu_0 H_0, \omega_r=\sqrt{\omega_0(\omega_0+\omega_M)}$, $\Gamma=\alpha\omega, \alpha$ is the dimensionless Gilbert damping parameter, and γ is the gyromagnetic ratio for an electron. We have expressed the field and magnetization as frequencies in order to clarify the comparison to our frequency-dependent data. It is clear from Eq. (1) that for small damping the quantity ω_r is the ferromagnetic resonance frequency, and it can be shown that the antiresonance frequency, the point at which $\mu_1=0$, is given by $\omega_{ar}=\omega_0+\omega_M$. For simplicity, Eq. (1) has been written for the limiting case of an infinitely thin sample with the static magnetic field applied in the plane of the sample. For a finite sized sample there are corrections to this form due to demagnetization effects.[17]

As discussed above, the microwave reflection measurement which we have employed yields the surface impedance instead of the permeability, however the two are related as follows: $\hat{Z}_S(\omega,T)=\sqrt{i\omega\hat{\mu}(\omega,T)\rho}$. We have assumed that at microwave frequencies $La_{0.8}Sr_{0.2}MnO_3$ is in the Hagen-Rubens limit (i.e., $\rho_2\ll\rho_1\approx\rho_{dc}$), allowing us to insert a frequency-independent value for ρ. Then we can substitute the expression for the susceptibility from Eq. (1) into this expression in order to model the complete frequency dependence of R_S and X_S. Measurements of the surface impedance as a function of applied magnetic field have shown that this model provides an excellent description of the measured data.[18,19]

Figure 1 shows zero-field $R_S(\omega)$ and $X_S(\omega)$ spectra at various temperatures. The antiresonance features, minima in R_S, and steps in X_S are clearly visible at all temperatures below T_C, despite the fact that $H_0=0$. Naively we would expect the permeability to be dispersionless in the absence of a static magnetic field, and thus both $R_S(\omega)$ and $X_S(\omega)$ should have the square-root frequency dependence characteristic of a metal, as is seen in the 309 K spectra, above T_C. We can understand why this is not so if we consider that even in the absence of an *external* magnetic field, there are local internal fields H_i due to anisotropy and domain structure, for example. Although 180° domain walls separating magnetically saturated regions can in principle produce rather large internal fields,[17] our field-dependent measurements of the antiresonance as $H_0\rightarrow 0$ allow us to estimate that $\mu_0 H_i\leqslant 0.02$ T,[19] probably due to a more complicated domain structure. Such small but finite fields can, however, cause the precession of the magnetization and thereby the dispersion of the permeability. Since $\omega_{ar}=\gamma\mu_0(H_i+M_0)$ and $H_i\ll M_0$, we see a well-defined antiresonance feature at a frequency determined predominantly by M_0 and a width due to inhomogeneities in the internal fields and the intrinsic damping given by α. Therefore we can extract the temperature dependence of the magnetization, in the absense of an applied field, from the temperature dependence of the antiresonance frequency.

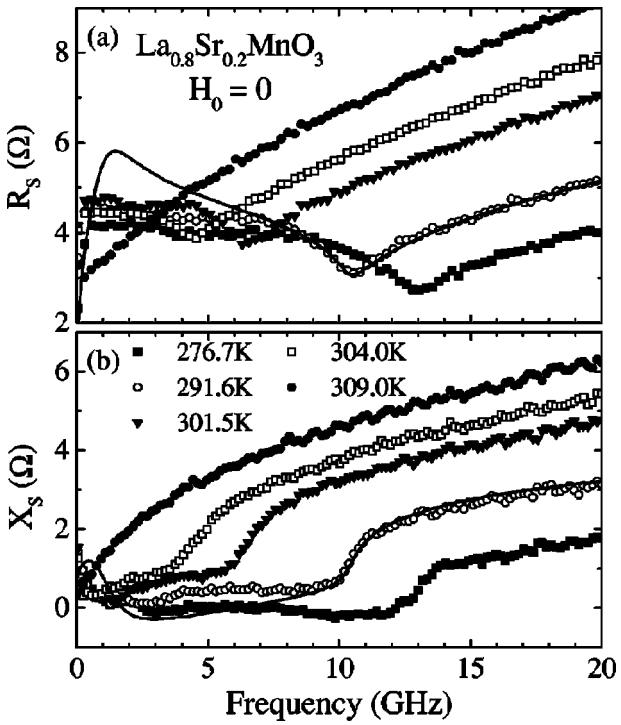

FIG. 1. The zero-field frequency dependence of (a) the surface resistance R_S and (b) the surface reactance X_S at various representative temperatures. The solid lines show a fit to the data at 291.6 K, as discussed in the text.

The solid lines in Fig. 1 are fits of the model presented above to the 291.6 K spectra, where we have set $H_0=0$, but included a finite damping to account for a distribution of internal fields around $H_i=0$. The discrepency between the model and the data at low frequencies is probably due to the fact that the model does not properly describe this distribution, but it is clear that the model does an excellent job of describing the behavior of both components of the surface impedance in the vicinity of the ferromagnetic antiresonance.

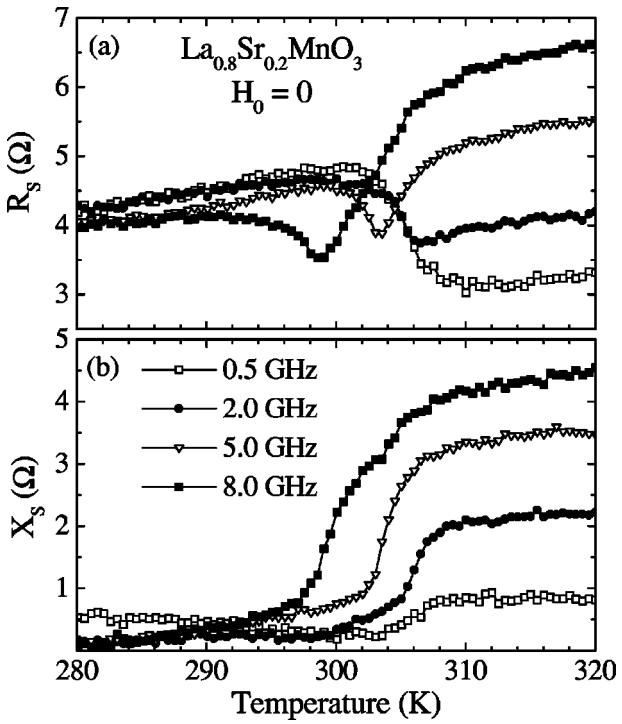

FIG. 2. The zero-field temperature dependence of (a) the surface resistance R_S and (b) the surface reactance X_S at various representative frequencies.

The surface impedance can also be measured as a function of temperature at fixed frequency, and Fig. 2 shows such data at a few representative frequencies. The antiresonance is manifested as a minimum in $R_S(T)$ coincident with an inflection point in $X_S(T)$, and moves to lower temperature as the frequency increases. It is, of course, also possible to extract $M_0(T)$ from these data, and therefore we have four sets of spectra from which to determine $M_0(T)$.

For the X_S data, both as a function of frequency and temperature, we have determined ω_{ar} by finding the peak of the first derivative, dX_S/df [Fig. 1(b)] or dX_S/dT [Fig. 2(b)]. Similarly, ω_{ar} is determined from the positions of the local minima in $R_S(f)$ and $R_S(T)$. Figure 3 shows the magnetization curve which we have extracted from these four sets of data. The onset of spontaneous magnetization at $T_C \approx 305.5$ K is clearly seen.

With these data it is possible to examine the critical behavior of the magnetization in the vicinity of T_C. The theory of critical phenomena at a second-order phase transition predicts that the magnetization will vary as a power law, $M_0(T) \propto (T_C - T)^{\beta}$, where β depends on the Hamiltonian describing the interactions among the spins.[12,13] Therefore, determination of this exponent yields information about the range of ferromagnetic interactions. But this expression is expected to hold only in the limit $T \to T_C^-$, so it is necessary to look at the behavior very close to T_C, where the slope of $M_0(T)$ is very large. One approach is to calculate the following function:[20] $T^*(T) = -M_0(dM_0/dT)^{-1} = \beta^{-1}(T_C - T)$. Thus T^* is linear in T, and values of β and T_C can be determined from the slope and intercept. For the data shown in Fig. 3 we find that T^* is roughly linear for temperatures above 290 K, and below this temperature the slope of $T^*(T)$ increases due to the saturation of the magnetization. It is

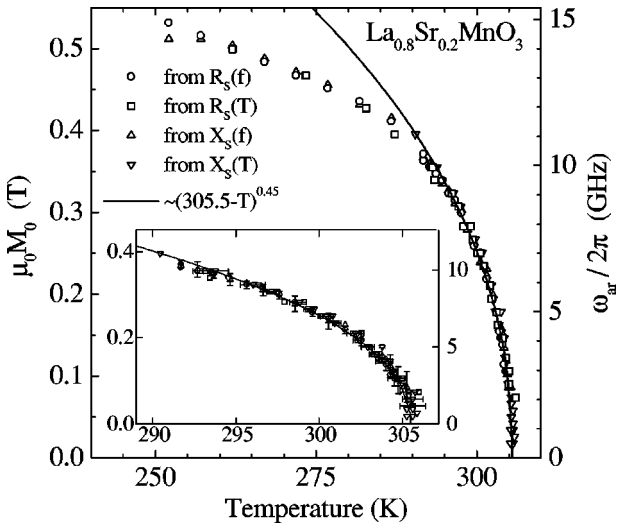

FIG. 3. The spontaneous magnetization M_0 of $La_{0.8}Sr_{0.2}MnO_3$ extracted from the four sets of data shown in Figs. 1 and 2 (for each data set there are many more spectra than are shown in these figures). The inset shows the same data over a smaller temperature range near T_C, and the solid lines are a fit to the data with the scaling form given in the text. The right axes show the FMAR frequencies.

therefore only reasonable to examine the critical behavior in the temperature region between 290 K and T_C. If we define the dimensionless quantity $\varepsilon = 1 - T/T_C$ then we see that this corresponds to a range of $\varepsilon \approx 0 - 0.05$. By fitting a straight line to this portion of the $T^*(T)$ curve we find that $T_C = 305.5 \pm 0.5$ K and $\beta = 0.45 \pm 0.05$. The solid line in Fig. 3 shows the critical behavior with these parameters, and the inset shows the same data and model between 290 K and T_C, with some representative error bars included.

A previous study of the microwave properties of similar $La_{0.8}Sr_{0.2}MnO_3$ single crystals gave values of $T_C = 304 \pm 3$ K and $\beta = 0.34 \pm 0.05$.[7] This value of β is clearly in disagreement with ours, even though the $M_0(T)$ data from the two experiments are in agreement. This apparent contradiction can be understood as follows: in the earlier study, the value of β was obtained by fitting the magnetization data between 270 and 300 K ($\varepsilon \approx 0.01 - 0.11$). As we have shown above, the use of data below 290 K is not appropriate for an examination of the critical behavior. In addition, our ability to measure FMAR to lower frequencies allows us to determine the magnetization much closer to T_C, which is the most important region for an accurate determination of both β and T_C. If we fit our data only over the range $\varepsilon \approx 0.01 - 0.11$, then we find $T_C = 303$ K and $\beta = 0.26$. If instead we fix T_C at 305.5 K and allow only β to vary, again fitting over the same range, we find $\beta = 0.33$. So it is clear that the difference between our result and that of the previous measurement arises simply from the temperature range over which the analysis of the critical behavior has been done.

Our experimental value of the scaling exponent β is in agreement with mean-field theory, which predicts $\beta = 0.5$.[12,13] Similar results were obtained from microwave measurements on $La_{0.7}Sr_{0.3}MnO_3$ single crystals, which gave $\beta = 0.45 \pm 0.05$.[6] Their fit range was $\varepsilon \approx 0.01 - 0.12$. And recently reported dc magnetization measurements on polycrystalline $La_{0.8}Sr_{0.2}MnO_3$, also gave $\beta = 0.50 \pm 0.02$, using a

narrow temperature range of $\varepsilon \approx 0-0.01$.[9] However, neutron-scattering and dc magnetization measurements on $La_{0.7}Sr_{0.3}MnO_3$ single crystals yielded $\beta = 0.295 \pm 0.002$ ($\varepsilon \approx 0-0.13$) and $\beta = 0.37 \pm 0.04$ ($\varepsilon \approx 0-0.03$), respectively.[4,11] Recent neutron-scattering measurements on single crystals of both $La_{0.8}Sr_{0.2}MnO_3$ and $La_{0.7}Sr_{0.3}MnO_3$ gave values of $\beta = 0.29 \pm 0.01$ ($\varepsilon \approx 0-0.18$) and $\beta = 0.30 \pm 0.02$ ($\varepsilon \approx 0-0.3$), respectively.[10] Finally, a measurement of the temperature dependence of the zero-field muon precession frequency in $La_{0.67}Ca_{0.33}MnO_3$ gave $\beta = 0.345 \pm 0.015$ ($\varepsilon \approx 0.03-0.27$).[5] Although many of these studies yielded values of β which are significantly lower than what we have found, the explanation for this seems to lie in the fact that the saturation of the magnetization will always lead to a reduced value for β if the data are fit over too wide a temperature range.

In conclusion, we have extracted the zero-field spontaneous magnetization of single-crystal $La_{0.8}Sr_{0.2}MnO_3$ from the temperature and frequency dependence of the microwave surface impedance. This magnetization rises continuously below T_C, as expected for a second-order phase transition. Analysis of these data gives values for the Curie temperature $T_C = 305.5 \pm 0.5$ K and the scaling exponent $\beta = 0.45 \pm 0.05$. Unfortunately, it seems that there is not yet experimental consensus about the scaling behavior of the magnetization in these compounds. It is clear that the value of β is very sensitive to the fit range, with values ranging from $\beta = 0.3-0.5$ reported in the literature. The technique presented here is unique because it requires no external field and is broadband, both of which allow us to examine the asymptotic critical behavior of the magnetization as $T \rightarrow T_C$. The result presented here is consistent with the mean-field value (0.5), implying that there are long-range ferromagnetic interactions in this lanthanum manganite.

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