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1997 J. Phys.: Condens. Matter 9 L633

(http://iopscience.iop.org/0953-8984/9/49/001)

J. Phys.: Condens. Matter **9** (1997) L633–L639. Printed in the UK PII: S0953-8984(97)88257-4

LETTER TO THE EDITOR

Temperature-tuned natural ferromagnetic resonances in $La_{0.9}Sr_{0.1}MnO_3$

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Received 2 October 1997

Abstract. We present the results of an investigation into the zero-field microwave absorption in $La_{0.9}Sr_{0.1}MnO_3$ for frequencies ranging from 2 to 35 GHz. The temperature dependence of the microwave loss at zero applied field displays two peaks for frequencies less than 15 GHz. These peaks can be ascribed to temperature tuning of the internal anisotropy field to satisfy the conditions for ferromagnetic resonance.

During the past year [1, 2] this laboratory has concentrated on using the temperature and field dependence of microwave absorption to elucidate the magnetic state of the manganite system $La_{1-x}Sr_xMnO_3$ with $x = 0.3$ and 0.2. Using high-quality single-crystal samples, it was shown (i) that the ferromagnetic transition is sharp, continuous and second order, (ii) that in the $x = 0.2$ case, there is a marked ($\sim$20 K) discrepancy between the Curie temperature T_C and the resistivity (ρ) peak temperature T_p; that is, $\partial\rho/\partial T$ changes sign while the material is a paramagnet. In addition to conventional measurements, we presented the novel technique of temperature-tuned (TT) ferromagnetic antiresonance (FMAR). In this method, one takes advantage of the rapid temperature variation of the magnetization, in the neighbourhood of T_C, to study the phenomenon of the FMAR dip in the microwave absorption [3] consequent upon a minimum in the dynamic permeability of a bulk metallic ferromagnet.

The present note concerns the temperature dependence of *zero-field* microwave losses in a single crystal of the dilute manganite $La_{0.9}Sr_{0.1}MnO_3$. Previous investigations [4–6] have shown that this material has an onset of ferromagnetism at about 150 K and a transition to a spin-canted state at about 110 K. Here it is found that the microwave absorption exhibits two large maxima as the temperature T is varied between 100 and 150 K. It is suggested that we have made the first observation of a temperature-tuned natural (zero-field) ferromagnetic resonance (TTFMR) phenomenon. The magnetocrystalline anisotropy implied by the TTFMR is in reasonable agreement with that derived from dc magnetization data taken on the *same* sample.

It is interesting to point out the analogy of the present experiment with the TT caliper resonance in a second-sound cavity containing superfluid helium [7] and the TT nuclear magnetic resonance in MnF_2 [8].

We will now describe the methods used. Single crystals of $La_{0.9}Sr_{0.1}MnO_3$ were grown by the floating-zone technique. X-ray diffraction studies revealed the samples to be single-phase crystals. A plate (2 mm $\times$ 2 mm $\times$ 0.2 mm) was cut and polished with its plane

0953-8984/97/490633+07$19.50

surface lying close to the *ab*-plane. As before, microwave absorption measurements were made using rectangular cavities operating at frequencies lying between 2 and 35 GHz. The sample was placed at the site of the maximal microwave magnetic field h_{rf}, and the absorbed power P was monitored between 77 and 300 K. In order to define T_C, the ac susceptibility was measured at 135 Hz for $4 < T < 300$ K, using a field amplitude of 10 Oe. The dc magnetization M of the same sample was obtained by SQUID magnetometry for temperatures between 5 and 300 K and applied fields H up to 50 kOe. Magnetic isotherms were taken both parallel and perpendicular to the sample plane. The dc magnetization data also exhibit several novel features at high fields. They will be presented elsewhere [9]. In this note we present just a few dc magnetization measurements to show that the magnetic easy axis is normal to the sample plane.

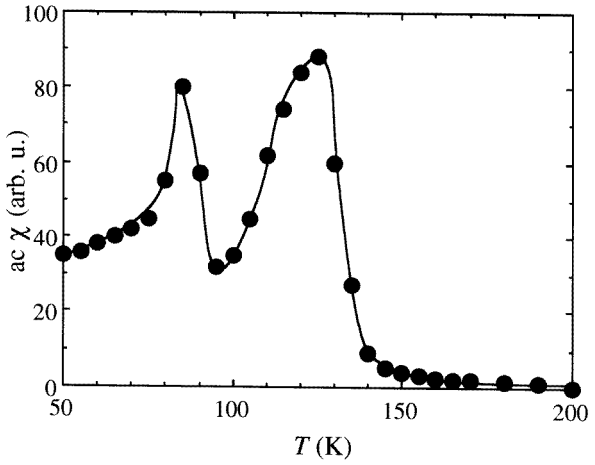

Figure 1. The temperature dependence of the real part of the ac susceptibility. The onset of ferromagnetism is near 150 K, and the presence of two maxima is suggestive of possible non-collinear magnetization.

We now describe our results. Figure 1 shows the ac susceptibility as a function of temperature and indicates a T_C of about 150 K, in agreement with other recent reports [4–6]. However, it is clear that there is a magnetic instability at low T. It is not possible to interpret the present result unequivocally. Whereas it is not inconsistent with the onset of spin canting [10], a sharp increase in the coercive field [11] could also account for the low-T behaviour shown in figure 1.

Treating the sample as a regular parallelepiped, one would estimate that the demagnetization factors N_x, N_y for $\boldsymbol{H}$ parallel to the sides are about $0.1 \times 4\pi$. The sum rule gives a demagnetization factor of $0.8 \times 4\pi$ along the normal. At 140 K (figure 2) magnetic isotherms in both the parallel and perpendicular geometries show saturation between 1.5 and 2 kOe. Along the easy axis, one expects saturation at $H = NM$, while along the hard direction, one would need $H = NM + H_k$ where H_k is the 'anisotropy field'. With a value of $4\pi M$ of about 2 kOe, figure 2 implies an anisotropy field of around 1.5 kOe with the symmetry (easy) axis pointing along the c-direction, i.e. normal to the sample plane.

Following the results of other investigators [4, 5], it is noted that the dc resistivity ρ is rather high ($\sim$1 Ω cm at 300 K) and increases monotonically on lowering T, with only a weak anomaly near 150 K. For the frequency range considered here, the sample can be considered to be an insulator as it is effectively transparent as far as the non-magnetic

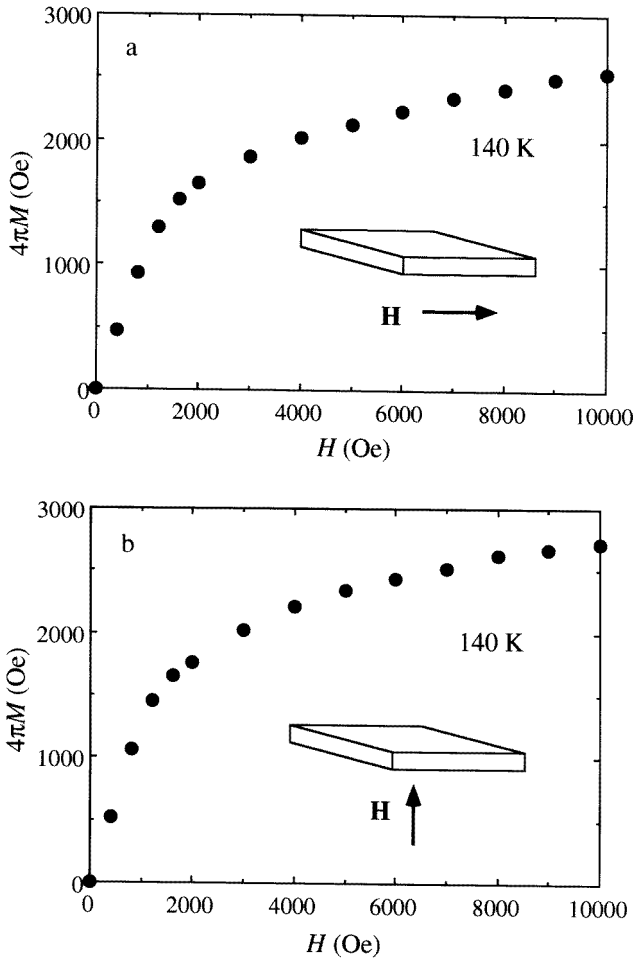

Figure 2. Magnetic isotherms at 140 K for $\boldsymbol{H}$ applied (a) in the sample plane, (b) along the sample normal. The in-plane magnetization saturates at nearly the same field as that out-of-plane, indicating the presence of an anisotropy field of about 1.5 to 2 kOe with an easy axis along the sample normal.

response is concerned.

In the light of the above, the T-dependence of the zero-field absorption P (figure 3) at 10 GHz comes as a complete surprise. Whereas between 300 and 150 K, P is indeed very small and insensitive to temperature, the loss *shows two large maxima* centred at about 137 and 102 K. The peaks are identified by the temperatures T_+ and T_-. One can also define a width ΔT_+ for the high-temperature peak which measures the temperature separation at half the maximum value of P. The temperature 'width' of T_- was considerably less precise. From the data listed in table 1, it can be seen that the peaks move lower in T and broaden as the frequency is raised. At still higher frequencies (>22 GHz), no peaks were discernible. As also seen in figure 3, for the lower frequencies, application of a field of 9 kOe completely suppresses the phenomenon. Finally, for T_-, there is thermal hysteresis; i.e. data taken while cooling and warming show different magnitudes for P and T_-.

We now discuss the results. The suppression of the maxima by the application of

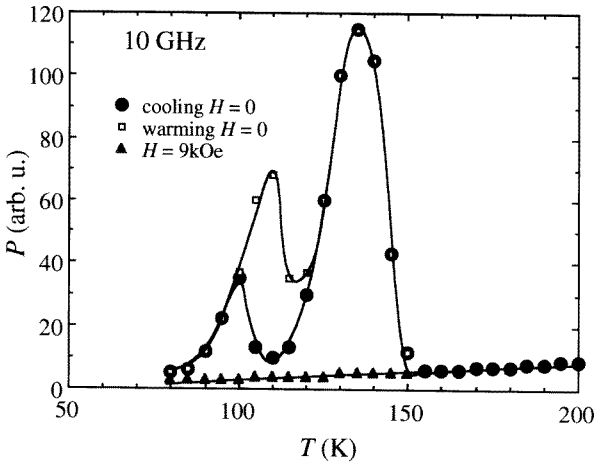

Figure 3. The temperature dependence of the microwave absorption at 10 GHz. At zero applied field, there are two peaks, and the lower peak displays thermal hysteresis. At a field of 9 kOe, the absorption is effectively absent.

Table 1. Characteristic temperatures for natural FMR in $La_{0.9}Sr_{0.1}MnO_3$.

Frequency (GHz)	T_+ (±3 K)	ΔT_+ (±5 K)	T_- (±5 K)
2.0	148	10	—
3.7	146	12	126
5.9	142	15	122
7.9	141	15	114
9.7	137	18	102, 110*
11.2	132	—	103, 109*
14.9	100	—	—

*Warming.

a magnetic field combined with the fact that the onset of the phenomenon occurs below 150 K (T_C) strongly suggests that the enhanced microwave absorption has a magnetic origin. Although the analysis is not exact for reasons which will be discussed, the present observations are attributed to TTFMR in the magnetocrystalline anisotropy. Since it appears in the absence of an applied field, following Smit and Wijn [12] we call this phenomenon natural FMR (NFMR).

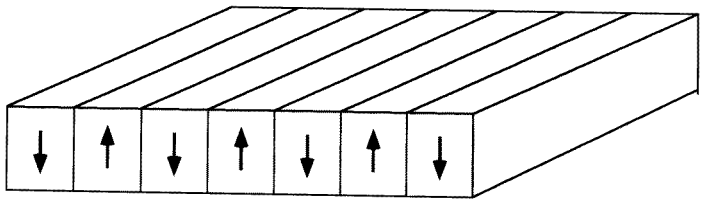

Figure 4. A schematic diagram of stripe domains in a thin film with the easy axis along the film normal. Note that for sufficiently thin films and close spacing, closure domains are not needed.

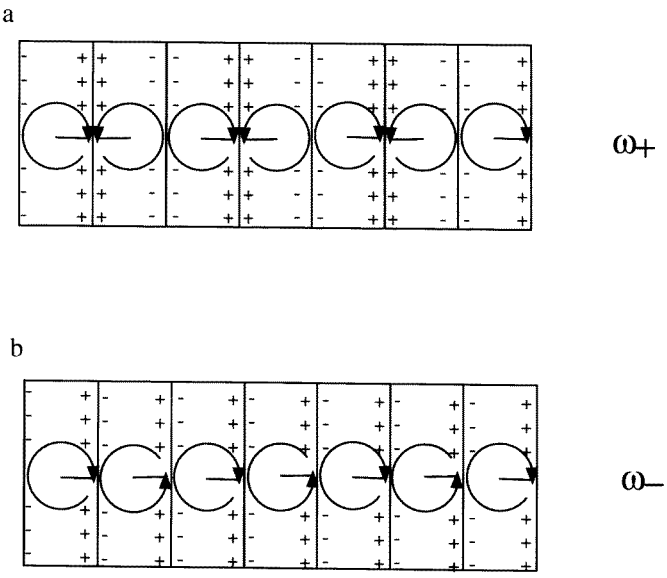

Figure 5. An overhead view of the precession of the spins in stripe domain patterns for (a) the ω_+-configuration and (b) the ω_--configuration. The symbols + and − represent the dynamic poles. In (a), the 'magnetic charges' at the wall interface add to each other, while in (b) they cancel.

At small (zero) applied fields, a ferromagnet (figure 4) spontaneously breaks up into domains in order to minimize the domain wall and demagnetization energies. For narrow stripe domains in sufficiently thin samples, with the spin direction normal to the sample plane, closure domains are not necessary. The present specimen is far from thin and therefore the details of the domain structure are likely to be complex. However, we believe that the essential physics is revealed by appeal to a simplified model—i.e. by introducing a uniaxial anisotropy K_u with the c-axis being the symmetry axis. The spins in each domain will precess in the anisotropy field $H_k = 2K_u/M$. The application of a radio-frequency magnetic field normal to the direction of the domain magnetization causes the spins to exhibit resonant absorption. In neighbouring domains (figure 5), the magnetizations are antiparallel and the spins precess in opposite directions. As first shown by Smit and Beljers [13] for stripe domains (and ignoring closure domains), there are two possible dynamic configurations. In neighbouring domains, the components of the magnetization which are normal to the domain wall either meet at the domain wall (figure 5(a)) or are 180° out of phase (figure 5(b)). For the former case (figure 5(a)), the spins create a dynamic demagnetizing field with the result being that the precession frequency becomes

$$\omega_+ = \gamma[H_k(H_k + 4\pi M)]^{1/2} \tag{1}$$

where $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio. For the other situation (figure 5(b)), the dynamic magnetic poles cancel each other, and the only torque on the spins is from H_k. One finds that in this case NFMR will occur when

$$\omega_- = \gamma H_k. \tag{2}$$

The designations ω_- and ω_+ reflect the fact that $\omega_+ > \omega_-$. Indeed, frequency-swept measurements [14, 15] on various hexagonal ferrites at $H = 0$ indicate two resonances.

In principle, equations (1) and (2) could be modified to take account of various domain structures and magnetocrystalline anisotropies. However, there are not enough data to warrant such an exercise.

In contrast to previous investigations of NFMR, in the present experiments the temperature was varied while holding ω constant. As T is lowered from T_C, one can expect to observe one or two NFMR, depending upon whether equation (1) or equation (2) or both are satisfied. Also, if the chosen frequency is too high it may not be possible to observe any peak. Furthermore, the ω_+ NFMR should appear at a higher temperature.

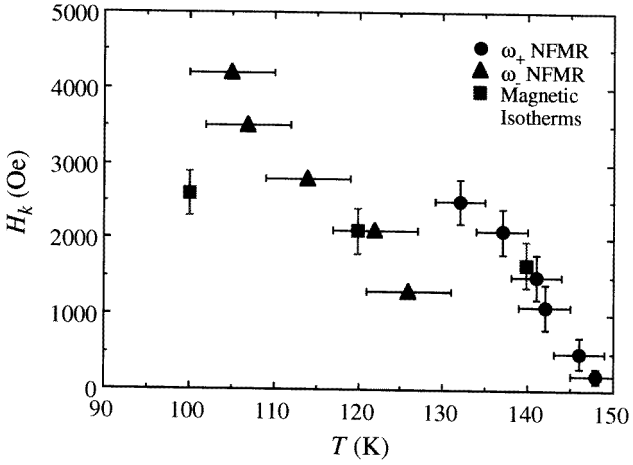

Figure 6. The temperature dependence of H_k deduced from magnetic isotherms and the ω_+ and ω_- NFMR conditions. Proper account of both the static and dynamic demagnetization factors would bring the NFMR values into better accord.

It seems reasonable to claim that the absorption maxima shown in figure 3 represent ω_+ and ω_- NFMRs in the magnetocrystalline anisotropy field of the *ab*-plane parallelepiped. If so, we can immediately obtain the relevant NFMR parameters. Figure 6 displays the temperature dependence of H_k as deduced from the magnetic isotherms as well as using equations (1) and (2) at T_+ and T_-, respectively. We assume a g-value of 2, following our electron spin-resonance results [16] on this material, and the spontaneous magnetization $4\pi M(0)$ was accessed by extrapolating the high-field isotherm data to zero field. One finds rough agreement among the three methods. It is worth noting that in equations (1) and (2), firstly, no account has been taken of any static demagnetizing field along the direction of spin alignment, and, secondly, it has been assumed that in the ω_--mode the cancellation is exact. A proper accounting for these effects would lower the derived H_k for ω_+ while raising that of ω_-, bringing the values closer together.

The increase of ΔT_+ (table 1) with increasing frequency is partly a result of the fact that at lower T, the temperature variation of H_k is slower. Although one might have expected smaller values of ΔT_+ at the lowest frequencies (highest T), the observed widths at low frequency may arise from a distribution of T_C-values in the sample, a not uncommon occurrence in the manganites [17].

In conclusion, we have studied the zero-field microwave absorption in a single crystal of $La_{0.9}Sr_{0.1}MnO_3$. For low enough frequency, P exhibits two peaks when T is lowered below T_C. The phenomenon can be explained by appeal to a temperature-tuned natural ferromagnetic resonance which arises from the internal field created by the

magnetocrystalline anisotropy. This is the first reported observation of either zero-field FMR in a manganite or a temperature-tuned natural FMR.

We thank S D Tyagi and P Lubitz for fruitful discussion. This research was supported in part by the University of Maryland NSF MRSEC.

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