PHYSICAL REVIEW B **69**, 100409(R) (2004)

Microwave absorption in a thin $La_{0.7}Sr_{0.3}MnO_3$ film: Manifestation of colossal magnetoresistance

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(Received 22 December 2003; published 19 March 2004)

Microwave (MW) absorption by a thin $La_{0.7}Sr_{0.3}MnO_3$ film on a $SrTiO_3$ substrate is investigated at a 9.1 GHz as a function of a dc magnetic field. Features of this absorption, namely, the jump in the absorption derivative, have been detected as the applied field passes through its zero value. Hysteretic behavior of the jumps is also observed. The results are discussed based on the model in which MW losses, additional to the ferromagnetic resonance, arise due to attenuation of MW currents induced in the sample by both variable magnetic induction and MW electrical field near the substrate surface with high dielectric permittivity. We show that zero-field anomalies in MW absorption are directly coupled with manganite magnetoresistive properties.

DOI: 10.1103/PhysRevB.69.100409 PACS number(s): 75.70.−i, 75.47.Gk, 76.50.+g

Electron magnetic resonance (EMR), namely, ferromagnetic resonance (FMR) at temperatures (T) below the Curie point T_C, and electron paramagnetic resonance (EPR) at $T > T_C$ have been extensively used for the study of doped manganite oxides. For example, the surface impedance in the range of the metal-insulator transition was studied in the works of Lofland *et al.*[1–4] EMR measurements were employed for studying the spin structure and magnetic (in)homogeneities in bulk and thin-film samples (see, e.g., Refs. 5–8), too. Even though the data on EMR in conductive magnetic materials can be influenced by eddy current losses of a magnetic component of the microwave (MW) field,[9,10] investigations[1–8] do not show any manifestation of the colossal magnetoresistivity (CMR) of manganites in the MW region. The MW absorption of thin $La_{1-x}Ba_xMnO_{3-z}$ films with a linear dependence on the modulus of the external magnetic (H) field was observed in Ref. 11. The authors interpreted the data as a manifestation of the MW CMR of the studied films; however, the physics behind the phenomena was not considered.

In this paper, the losses at frequency $\sim 10^{10}\ s^{-1}$ in the thin $La_{0.7}Sr_{0.3}MnO_3$ film on the $SrTiO_3$ substrate are investigated as a function of the applied H field. New anomalies in the MW absorption of the manganite film at the low magnetic field, which to a certain extent are equivalent to those observed in Ref. 11 but reveal new aspects of it, have been observed. The anomalies, namely, the jumps in the absorption derivative on the magnetic field at the field changing the sample magnetization sign are directly connected with the CMR of manganites that is manifested at this frequency. The hysteretic behavior is a representative feature of the jump as well. The results can be explained taking into account the Joule losses related both to FMR in a conducting ferromagnet and the MW electrical (**e**) field on the substrate surface with high permittivity ε. The condition when the anomalies can be observed is also discussed.

Experiments were performed on the $2.5 \times 4.0\ mm^2$ film with $\sim 0.3\ \mu m$ thickness grown on the 0.3 mm (001)-oriented $SrTiO_3$ substrate. The sample was placed at the MW magnetic field loop of TE102 cavity of the X-band EPR spectrometer Radiopan SE/X-2544. The temperature was in the range $100 \div 400$ (± 0.5) K. The accuracy of the sample to the H-field direction orientation was about $\pm 2°$. The investigated film resistivity achieves its maximum at ≈ 320 K and drops down with decreasing temperature, i.e., it shows a temperature dependence typical for such materials. The magnetoresistive effect, $[R(H=0) - R(H=1T)]/R(H=0) = 25\%$ at 310 K, is a representative one, too.

At $T \geqslant 360$ K a rather narrow isotropic EPR signal with a g factor $g = 1.97 \pm 0.01$ was observed. The peak-to-peak linewidth $\mu_0 \Delta H_{pp}$ was about 20 mT (μ_0 is the permeability of free space). At lower temperatures we observed a dependence of the resonance line position on the angle θ between the film's normal and H-field direction **n**, which further transforms at $T < T_C \approx 316$ K into a typical dependence for the FMR line of a film with the "easy plane" type anisotropy. Figure 1 shows the FMR field temperature behavior for parallel and perpendicular film plane geometries. They are described well by the usual Kittel's equations.[12] A comparison of Fig. 1 data with that known from the literature (see, e.g., Refs. 3 and 13) shows that the film's anisotropy is mainly determined by the sample shape (the demagnetization factor anisotropy). In our experiment, at least one of the substrate linear size, d_i, fulfills the relation $d_i \sqrt{\varepsilon} \geqslant n\lambda/2$, where λ is the MW wavelength in air and n is an integer. Due to the temperature dependence of ε some dielectric resonator modes can be equal to the measured frequencies at appropriate narrow temperature areas. Its influence on the registered spectra can also be seen (see the features at ≈ 145 and 195 K, and others in Fig. 1).

The representative records of the FMR spectra for the derivative of the MW absorption on a field, $dP(H)/dH$, obtained at $T = 298$ K and different angles θ are shown in Fig. 2 for the MW magnetic field of the cavity, $\mathbf{h}_{MW}$, in the film plane. The jumps on the spectra at the zero-field region are clearly seen. Except for the main FMR line, one can also

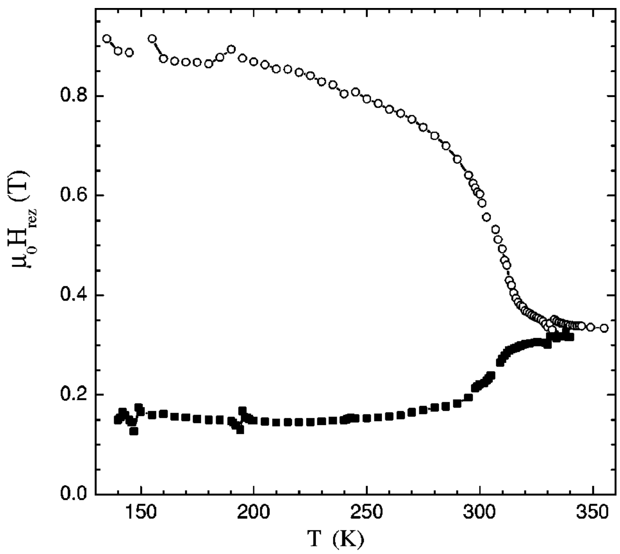

FIG. 1. Temperature dependence of the FMR resonant fields for longitudinal, $\theta = 90°$ (solid symbols), and transversal, $\theta = 0°$ (open symbols), magnetization. The features on FMR curves at ≈ 145, 195, 240, and 335 K are due to a coincidence of the measurement frequency and the dielectric resonator modes.

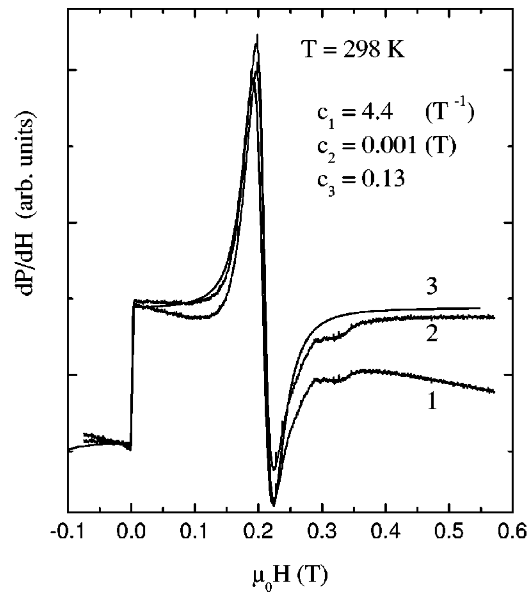

FIG. 3. The FMR spectrum at $T = 298$ K for $\theta = 90°$, $\mathbf{n} \perp \mathbf{h}_{MW}$. Line 1, spectrum "as it recorded;" line 2, the baseline corrected "as it seems more correct to the experimenter;" line 3, fitting by the function (3) with $c_1 = 4.4T^{-1}$, $c_2 = 0.001T$, and $c_3 = 0.13$.

observe well-resolved bulk nonuniform spin-wave modes on the low-field side and surface modes on the high-field side. These modes are quite similar to those investigated earlier on $La_{0.7}Mn_{1.3}O_3$ films.[14]

The observation of an unusual jump in the $dP(H)/dH$, which we detect sweeping the field through zero value, is the main result of our paper. The jump is very sharp for the "field-in-plane" geometry ($\theta = 90°$), while for $\theta \rightarrow 0$ it is smoother and its height is decreased. The dependence of the jump height on θ is shown in the inset of Fig. 2; it is close to $\sin\theta$ dependency. In detail the typical $dP(H)/dH$ dependence vs field in plane is shown in Fig. 3. The absorption curve $P(H)$, obtained by numerical integration of the Fig. 3 spectrum, is shown in Fig. 4. Note that the result of integration depends on the assumption about the baseline behavior in a high-field region. In Fig. 4 we present the results for different baseline asymptotic behaviors, to be confident that the zero-field anomaly is not sensitive to them. As is seen, the jump in the $dP(H)/dH$ dependence corresponds to a sharp turn in the dependence $P(H)$ at $H \approx 0$. For the spectrum in Fig. 3, the ratio of the jump's height to the absorption derivative swing in the region of the FMR line is as large as

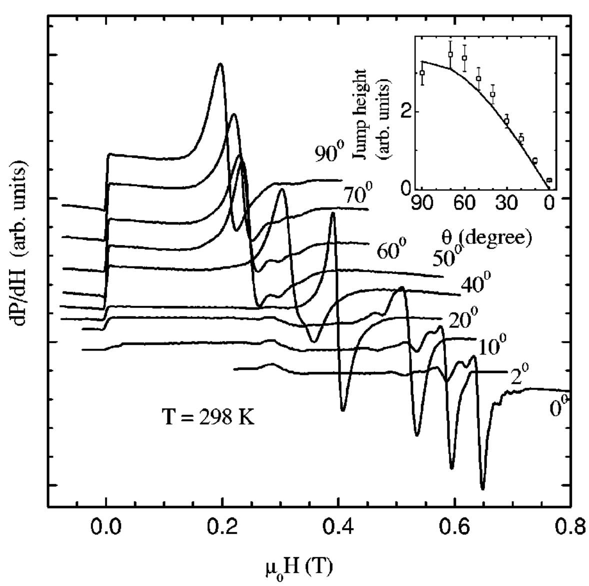

FIG. 2. The FMR spectra at $T = 298$ K for different orientations of the magnetic field ($\mathbf{n} \perp \mathbf{h}_{MW}$). Inset: the θ dependence of the jump height; points—the experimental values; the solid line is the $\sin\theta$ dependence.

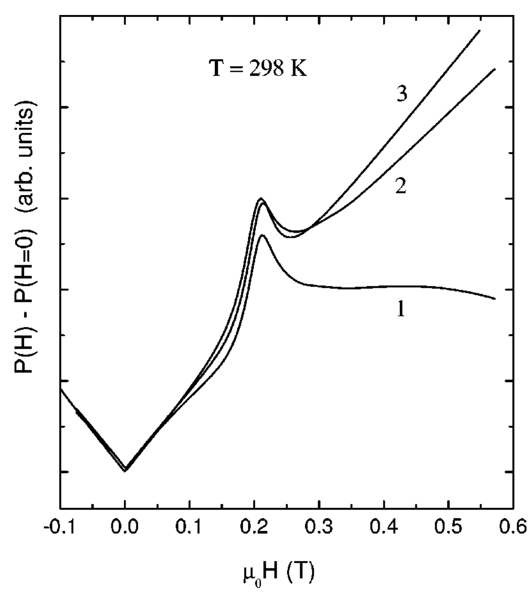

FIG. 4. The FMR absorption spectrum at $T = 298$ K for $\theta = 90°$ obtained by numerical integration of the data in Fig. 3. Lines 1, 2, and 3 are the same as in Fig. 3.

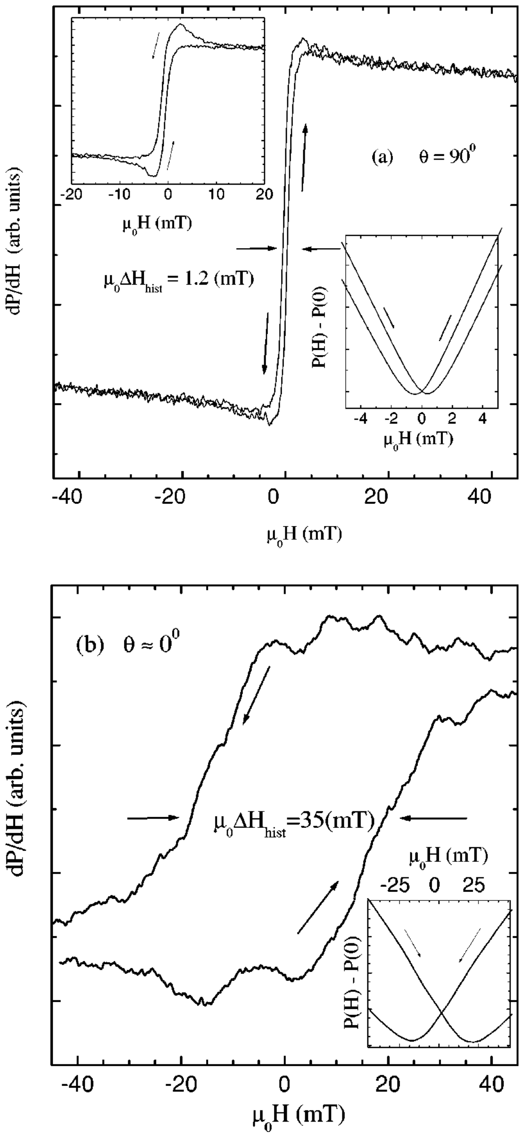

FIG. 5. The hysteretic loop of the absorption derivative jump at $T=298$ K, $\mathbf{n}\perp\mathbf{h}_{MW}$ for the magnetic field (a) in-plane and (b) close to normal to the film. Bottom insets: the part of the hysteretic loop obtained by integration of the derivative spectrum. Top inset: the absorption derivative of the jump hysteretic loop at a temperature where the dielectric resonator mode is close to the measurement frequency.

0.33. In the geometry $\mathbf{H}\perp\mathbf{n}\|\mathbf{h}_{MW}$ (not shown) this ratio gets the value 0.78.

The sudden change in the $dP(H)/dH$ dependence takes place not precisely at $H=0$ but is a little bit shifted from this point in the direction of the field sweeping. As a result, the records of two sweepings in opposite directions give almost a rectangular hysteretic loop. The example of such loops is shown in Figs. 5(a) and 5(b) for the ''field-in-plane'' and close to ''field normal-to-plane'' geometry, respectively. The angular dependence of the hysteretic loop width ΔH_{hist} is very well described by the relation $\mu_0\Delta H_{hist}\approx 1.4/\sin\theta$ mT.

Any nonlinear effects were not detected in the testing measurements at different (up to 15 dB) values of MW power submitted to the sample. So, the main distinguishing features of the anomalous MW absorption of the manganite film at low magnetic field are as follows: (i) the absorption is linear on the modulus of the H field; (ii) the $dP(H)/dH$ behavior is hysteretic; (iii) the angular behavior of the $dP(H)/dH$ jump magnitude is close to $\propto\sin\theta$, while the hysteretic loop width vs the field direction follows the $(\sin\theta)^{-1}$ dependence; and (iv) at given θ the jump relative height depends on measurement geometry.

There are a few possible physical mechanisms behind the anomalous jumps of $dP(H)/dH$ near zero field. Particularly, one can expect a jump every time when the field rearranges the magnetic domains if the absorption linewidth is comparable with the resonant H-field value (assuming the rectangular hysteretic loop). The height of such a jump should be about twice the tail of the absorption derivative line in the reorientation region. Therefore, for the FMR spectrum given in Fig. 5(a), with $\mu_0H_{rez}=210$ mT and $\mu_0\Delta H_{pp}=28.9$ mT, the magnitude of the jump should be about 0.004 of the absorption derivative swing of the FMR line, but the experimentally observed value is much larger (0.33). So, this mechanism is not the case.

Additional FMR anomalous absorption can be explained taking into account the Joule losses in the film. While the sample is placed in the cavity region without the MW **e**-field component, the MW **e** field, $\mathbf{e}_{ac,ind}$, is induced by the MW magnetic induction, $\mathbf{b}_{ac}=\mu_0(\mathbf{h}_{ac}+4\pi\mathbf{m}_{ac})$ and, in its turn, causes the eddy current losses. Here $\mathbf{h}_{ac}$ is the MW magnetic field in the sample, which takes into account the demagnetization factor, and $\mathbf{m}_{ac}$ is the ac magnetization. The field $\mathbf{e}_{ac,ind}$ depends on the sample shape and size,[9,10] on the H-field value and the FMR linewidth. The latter dependence appears through the usual relation $\mathbf{m}_{ac}=\chi_{ac}\mathbf{h}_{ac}$, where both the real χ'_{ac} and imaginary χ''_{ac} parts of magnetic susceptibility have well-known resonant dependence (see, e.g., Refs. 9 and 15). Also, if the external MW **e** field (e.g., **e** field of the cavity) $\mathbf{e}_{ac,ext}$ is present in the sample, it must be taken into account as well, and then the total MW **e** field acting on the sample is $\mathbf{e}_{ac}=\mathbf{e}_{ac,ind}+\mathbf{e}_{ac,ext}$.

The total MW absorption of the sample can be written as $P_{tot}=P_{FMR}+P_\sigma$, where $P_{FMR}=0.5\omega_{rez}\chi''_{ac}Vh^2_{ac}$. The Joule losses P_σ are proportional to $\mathbf{e}^2_{ac}$, the sample's conductivity $\sigma(T,H)$, and can be written in the form

$$P_\sigma=\tfrac{1}{2}\sigma(T,H)\int_V \mathbf{e}^2_{ac}dV=P(\mathbf{e}_{ac,ind})+P(\mathbf{e}_{ac,ext}). \qquad (1)$$

According to Refs. 9 and 10, the losses $P(\mathbf{e}_{ac,ind})$ and P_{FMR} can be summarized in the form $\tilde{P}_{FMR}=0.5\omega_{rez}\tilde{\chi}''_{ac}Vh^2_{ac}$, where $\tilde{\chi}''_{ac}$ corresponds to the usual expression for χ''_{ac} with a changed resonance half-width: $\delta H_{eff}=\Delta H_0+\delta_{ind}$. Here ΔH_0 is the initial FMR line half-width (without inclusion of the sample conductivity) and δ_{ind} is the correction to it connected with the additional damping of magnetization precession due to the field $\mathbf{e}_{ac,ind}$ and conductivity $\sigma(T,H)$. One can write approximately[9,10] $\delta_{ind}\approx\alpha_1\sigma(T,H)$, where α_1 is the parameter that depends on the sample form and size. Then the P_σ should be understood as $P(\mathbf{e}_{ac,ext})$ only; i.e., the additional nonresonance MW absorption is caused by the field $\mathbf{e}_{ac,ext}$ and is proportional to $\sigma(T,H)$.

As is known, the manganite material resistivity behavior on the H field can be phenomenologically described, assuming the dependence of the resistivity on $\mathbf{B}^2$, $\mathbf{BM}$, M^2, $|\mathbf{M}|$, etc., where $\mathbf{B}=\mu_0(\mathbf{H}+4\pi\mathbf{M})$. In particular, following Eq. (6) of Ref. 16, both the temperature and field dependencies of manganite magnetoresistivity can be qualitative described in terms of $|\mathbf{M}|$. Then, writing the magnetization as $\mathbf{M}(T,\mathbf{H})=\mathbf{M}_0(T)+\mathbf{H}\cdot[\partial\mathbf{M}(T)/\partial\mathbf{H}]|_{H\to 0+}+\cdots$ and taking into account that $[\partial\mathbf{M}(T)/\partial\mathbf{H}]|_{\mathbf{H}\to 0+}\approx\sin\theta\,\partial\mathbf{M}(T)/\partial\mathbf{H}_{\parallel\mathbf{M}}$ for the "easy-plane" film in the field $H\ll 4\pi M_0$, one can easily obtain for magnetoconductivity dependence on the field and angle

$$[\sigma(T,0)-\sigma(T,H)]\propto|H|\sin\theta=\sqrt{H^2}\sin\theta. \qquad (2)$$

Proceeding to Eq. (1), we obtain $P_\sigma\sim|H|\sin\theta$ in accordance with our data. Here $H\to 0+$ means that the derivative is taken on the same side of the region of magnetization reorientation, where $\sigma(T,H)$ is considered; $\partial\mathbf{M}(T)/\partial\mathbf{H}_{\parallel\mathbf{M}}$ is the $\mathbf{M}(T)$ derivative on the H field parallel to it. For the "easy plane" ferromagnet with the rectangular hysteresis loop and coercive field $H_{\mathrm{crc}}(T)$ the $[\sigma(T,0)-\sigma(T,H)]$ $\propto(|H_{\mathrm{crc}}(T)|-|H|\sin\theta)$ if $(|H_{\mathrm{crc}}|-|H|\sin\theta)>0$. In this case the $\sigma(T,H)$ dependence demonstrates hysteretic behavior with the loop width $\sim(\sin\theta)^{-1}$ in agreement with the measurements.

The fitting of the experimental data for $\theta=90°$ by the function

$$P_{\mathrm{fit}}(H)=\mathrm{const}\times\{c_1\sqrt{H^2}+\tilde{\chi}''_{\mathrm{ac}}(H,\delta H_{\mathrm{eff}})\}, \qquad (3)$$

with $\delta H_{\mathrm{eff}}=c_2+c_3\sqrt{H^2}$, is shown in Figs. 3 and 4. One can see a good agreement between the model and experimental data. The value of the fitting parameters $c_{1,2,3}$ and its physical origin will be discussed in detail elsewhere. Here we only note that the coefficient c_1 is responsible for the $dP(H)/dH$ jump and is caused solely by the MW **e**-field $\mathbf{e}_{\mathrm{ac,ext}}$ acting on the film.

What is the reason for the field presence if the sample is placed in the MW magnetic field loop of the cavity? In our opinion, the field $\mathbf{e}_{\mathrm{ac,ext}}$ appears to be due to large permittivity of the $SrTiO_3$ substrate. The **e** component of the MW field, being concentrated inside the substrate and penetrating partly into the manganite film, is responsible for the field $\mathbf{e}_{\mathrm{ac,ext}}$. It is worth noting that in Ref. 11 the manganite film was also placed in the region of the magnetic component of the MW cavity. Since the film in Ref. 11 had the underlying and cup $SrTiO_3$ layers, the field $\mathbf{e}_{\mathrm{ac,ext}}$ is also present on the sample. To test the idea, we repeated the measurements on the film of the same composition but on the $LaAlO_3$ substrate. Anomalies in the MW absorption were not observed. We also performed additional measurements with the sample shifted from the magnetic towards the electrical loop of the MW cavity. Now the ratio of the zero-field jump magnitude to the FMR $dP(H)/dH$ value does not depend on the shift, i.e., on the presence of the MW electrical field of the cavity on the sample. That is because, due to the wavelength difference, the uniform MW field of the cavity cannot excite the short electromagnetic wave in the substrate. It may take place only in the presence of heterogeneity with the appropriate size. The ferromagnetic film on the substrate is the main heterogeneity of the case and determines the condition for MW field excitation in the substrate by $\mathbf{h}_{\mathrm{MW}}$.

In conclusion, we report the preliminary results of CMR manifestation in the MW absorption by a thin $La_{0.7}Sr_{0.3}MnO_3$ film which is grown on the substrate with high permittivity and is placed in the magnetic loop of the cavity. Such features as the jumps in the absorption derivative on the H field and the hysteretic behavior of the jumps have been detected as the applied field passes through its zero value. The additional losses arise due to the damping of the eddy currents created in the manganite film by the MW **e** field. This field is inducted by the magnetization excited in the FMR process and by the penetration of the electrical MW field components from a high-dielectric substrate into the magnetic film.

[1] S. E. Lofland, S. M. Bhagat, H. L. Ju, G. C. Xiong, T. Venkatesan, R. L. Greene, and S. Tyagi, J. Appl. Phys. **79**, 5166 (1996).

[2] S. E. Lofland, P. H. Kim, P. Dahiroc, S. M. Bhagat, S. D. Tyagi, C. Kwon, R. Shreekala, R. Ramesh, and T. Venkatesan, J. Phys.: Condens. Matter **9**, 6697 (1997).

[3] S. E. Lofland, V. Ray, P. H. Kim, S. M. Bhagat, M. A. Manheimer, and S. D. Tyagi, Phys. Rev. B **55**, 2749 (1997).

[4] S. E. Lofland, S. M. Bhagat, H. L. Ju, G. C. Xiong, T. Venkatesan, and R. L. Greene, Phys. Rev. B **52**, 15 058 (1995).

[5] A. I. Shames, E. Rosenberg, G. Gorodetsky, W. H. McCarroll and M. Greenblatt, Mater. Sci. Forum **373-378**, 63 (2001).

[6] A. Rinkevich, A. Nossov, V. Vassiliev, and V. Ustinov, Phys. Status Solidi A **179**, 221 (2000).

[7] V. V. Srinivasu, S. E. Lofland, and S. M. Bhagat, J. Appl. Phys. **83**, 2866 (1998).

[8] V. V. Srinivasu, S. E. Lofland, S. M. Bhagat, K. Ghosh, and S. D. Tyagi, J. Appl. Phys. **86**, 1067 (1999).

[9] A. G. Gurevich, Fiz. Tverd. Tela (Leningrad) **16**, 1784 (1974) [Sov. Phys. Solid State **16**, 1159 (1974)]; A. G. Gurevich, *Magnetic Resonance in the Ferrites and Antiferromagnetics* (Nauka, Moscow, 1973).

[10] M. Marysko, Phys. Status Solidi A **28**, K159 (1975).

[11] S. D. Tyagi, S. E. Lofland, M. Dominguez, S. M. Bhagata, C. Kwon, M. C. Robson, R. Ramesh, and T. Venkatesan, Appl. Phys. Lett. **68**, 2893 (1996).

[12] C. Kittel, Phys. Rev. **71**, 270 (1947).

[13] S. E. Lofland, S. M. Bhagat, K. Ghosh, R. L. Greene, S. G. Karabashev, D. A. Shulyatev, A. A. Arsenov, and Y. Mukovskii, Phys. Rev. B **56**, 13 705 (1997).

[14] V. Dyakonov, A. Prohorov, V. Shapovalov, V. Krivoruchko, V. Pashchenko, E. Zubov, V. Mihailov, P. Aleshkevych, M. Berkowski, S. Piechota, and H. Szymczak, J. Phys.: Condens. Matter **13**, 4049 (2001).

[15] B. Lax and K. J. Button, *Microwave Ferrites and Ferrimagnetics* (McGraw-Hill, New York, 1962).

[16] J. O'Donnell, M. Onellion, M. S. Rzchowski, J. N. Eckstein, and I. Bozovic, Phys. Rev. B **54**, R6841 (1996).