Giant radio-frequency magnetoabsorption effect in the cobaltite ceramic La$_{0.5}$Sr$_{0.5}$CoO$_3$

B. I. Belevtsev,¹† A. Ya. Kirichenko,² N. T. Cherpak,² G. V. Golubnichaya,² I. G. Maximchuk,² A. B. Beznosov,¹ V. B. Krasovitsky,¹ P. P. Pal-Val,¹ and I. N. Chukanova³

¹B. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Sciences, Kharkov 61102, Ukraine
²A. Uskov Institute for Radiophysics and Electronics, National Academy of Sciences, Kharkov 61085, Ukraine
³Institute for Single Crystals, National Academy of Sciences, Kharkov 61001, Ukraine

The DC transport properties of and the radio-frequency (RF) wave absorption (at 1.33 MHz) in a ceramic sample of La$_{0.5}$Sr$_{0.5}$CoO$_3$−δ are measured. The Curie temperature, $T_c$, of the sample is about 250 K. A giant negative magnetoabsorption effect is found. In the vicinity of $T_c$, the absolute value of the magnetoabsorption is about 38% in the rather low magnetic field 2.1 kOe. This differs drastically from the measured DC magnetoresistance (MR) $\delta(H) = |R(0) - R(H)|/R(0)$ which is a mere 0.26% near $T_c$ in the same field and increases to about 2.15% in $H = 20$ kOe. The phenomenon can be understood taking into account that the magnetoabsorption is determined by influence of magnetic field on the conductivity and the magnetic permeability, while the MR is determined solely by the former. The magnetoabsorption effect can be used to develop RF devices controlled by magnetic field and temperature.

PACS numbers: 72.80.Ga; 75.30.Vn; 78.70.Gd

I. INTRODUCTION

In recent years much attention has been paid to mixed-valence cobaltites of the type La$_{1-x}$A$_x$CoO$_3$−δ ($0 < x \leq 0.5$). Although the system was studied for more than 50 years, not a few of its unique magnetic and transport properties are still puzzling. The parent compound LaCoO$_3$ is a non-magnetic insulator. For the doping range $0.3 < x \leq 0.5$, the system is ferromagnetic (FM) with the Curie temperature, $T_c$, in the range 220−250 K. It is known that absorption of electromagnetic waves is a non-magnetic insulator. For the doping range $0.3 < x \leq 0.5$, the system is ferromagnetic (FM) with the Curie temperature, $T_c$, in the range 220−250 K.

It should be pointed out that much more interest is being shown in related FM perovskite-like oxides, mixed-valence manganites, of the type La$_{1-2x}$A$_x$MnO$_3$, where A is a divalent alkaline-earth element like Ca, Sr, Ba. In these compounds, the effect of so-called colossal magnetoresistance (CMR) is found which offers applications in advanced technology. It is usual, in this connection, when considering properties of the cobaltites to compare them with those of the manganites. The magnetoresistance (MR) in manganites, defined as $\delta(H) = |R(0) - R(H)|/R(0)$, is found to be more than 90% at a field $H$ of about 60 kOe in the neighbourhood of room temperature. In spite of enormous efforts, a clear understanding of CMR is not yet available, though some models describe experimental data with an acceptable accuracy. Beside this, the requirement of high magnetic fields hampers the possible application of the CMR manganites. In the Sr-doped cobaltites, the magnitude of the MR is found to be rather low (only a few per cent). It is hoped that elucidation of large difference in the MR between the manganites and cobaltites could be helpful for understanding the nature of CMR.

The study presented in this article is inspired to a certain extent by results of Ref. 10, where transport properties in a bulk ceramic sample and a film of La$_{0.5}$Sr$_{0.5}$CoO$_3$−δ were measured by DC and microwave (41 GHz) methods. It was found that the film presents a highly inhomogeneous system of weakly connected FM grains. The most important finding in Ref. 10 is the following: the microwave conductivity of the film, which should be related mainly to the conductivity within the poorly connected grains, increases by an order of magnitude at the transition to the FM state. This change is huge in comparison with the 10% decrease in the film DC resistivity found in the same temperature range. The main conclusion of Ref. 10 is that the microwave absorption reflects more closely the temperature variations in intragrain conductivity of inhomogeneous oxides than the DC conductivity.

It is known that absorption of electromagnetic waves in FM metals is determined not only by the conductivity, but by the magnetic permeability, $\mu$, of absorbing media as well. At a fairly high frequency, the magnitude of $\mu$ is close to unity. The second feature of the microwaves is their small penetration depth, which is called the skin depth. In good conductors, the skin depth of microwaves is a few micrometers. This is quite enough for thin films, but it often causes a problem in the study of bulk samples. The surface layer of bulk samples can have differing properties from the interior. This is especially true in the case of FM oxides (including cobaltites), where a surface is often depleted in oxygen. It is advisable, in this connection, to use low-frequency waves to study bulk samples to ensure a fairly large skin depth. But in this case the influence of permeability on the absorption of these waves should be expected.

†belevtsev@ilt.kharkov.ua
cherpak@ire.kharkov.ua
It follows from the results of Ref. [10] that high-frequency wave absorption might be much more sensitive to an applied magnetic field than DC resistivity. The aim of this study was to test this assumption. The subject of investigation was a bulk sample of La$_{0.5}$Sr$_{0.5}$CoO$_{3-δ}$. Radio-frequency (RF) waves penetrating most of the sample volume were used. We have found a giant magneetoabsorption (about 38%) near the Curie temperature in a rather small magnetic field 2.1 kOe. This differs drastically from the measured MR values (the $δ(H)$ is about 0.26 % in this field). The reasons for the giant magneetoabsorption effect are considered.

II. EXPERIMENTAL METHOD

The ceramic samples of La$_{0.5}$Sr$_{0.5}$CoO$_{3-δ}$ are all cut from the same pellet, prepared by a standard solid-state reaction technique. The pellet was polycrystalline with grain size in the range 40-70 μm. Some other details of the sample preparation and characterization can be found in Ref. [10]. The measurements of the AC permeability were done by an induction technique using a non-commercial magnetometer in AC magnetic field $H_{ac}$ = 4 Oe and frequency 10 kHz. The DC resistance, as a function of temperature and magnetic field $H$ (up to 20 kOe), was measured using a standard four-point probe technique.

The sample for RF measurements (with dimensions 21.6×7×4 mm$^3$) was placed in an induction coil (9.7 mm in diameter and with a height approximately equal to the longest side of the sample). This coil is a part of an LC tank circuit. The magnitude of RF magnetic field in the induction coil was not more than 0.1 Oe. The measurements of the quality factor, $Q$, of the LC tank with and without the sample inside the coil were taken. The measurements of the $Q$-factor depending on temperature or magnetic field were taken at fixed frequencies. In this article, we present the results for the frequency $ν$ = 1.33 MHz. The available cryostat with electromagnet makes it possible to measure the $Q$-factor in DC magnetic fields up to 2.5 kOe. The DC and RF fields were mutually perpendicular. The temperature and magnetic-field dependences of the $Q$-factor were recorded on heating after the sample had been cooled down in zero field.

Consider more exactly physical properties of the sample which can be derived or, at least, judged by from the $Q$-factor measurements of the RF LC tank employed. In the general case, the sample in an electromagnetic field can be described, using complex permittivity $ε_c = ε' - iε'' = ε'[1 - i tan(δ_ε)]$ (where $ε = ε_ε$, $tan(δ_ε) = σ_{ef}/(ωε)$ is the dielectric loss tangent, $σ_{ef}$ is the RF conductivity of the sample) and complex magnetic permeability $μ_c = μ' - iμ'' = μ'[1 - i tan(δ_μ)]$, where $μ = μ_0$ and $tan(δ_μ) = μ''/μ'$. For a plane electromagnetic wave, the complex resistance of the sample is given by relation

$$Z_c = \sqrt{\frac{μ_0c}{c_0}} = Z_0 \sqrt{\frac{μ_0}{ε_0} \left(\frac{1 - iμ''/μ'}{1 - iωτ_{rf}/ε_0ω}\right)} = R_c + iX_c,$$  

(1)

where $Z_0 = 120$ Ω, $R_c$ defines the energy loss in the sample, $X_c$ is the sample reactance.

Once the sample with the wave resistance $Z_c$ is inserted into the coil of the RF circuit, arranged from resistance, inductance and capacitance elements $R_0$, $L_0$ and $C_0$, a complex resistance is added to the circuit, given by

$$Z_{sp} = kZ_c = R_{sp} + iX_{sp},$$  

(2)

where $k$ is a geometrical factor determined by the sample and coil dimensions, which was about 0.38 in this study. The $Q$-factor of the circuit without the sample is expressed as $Q_0 = ωL_0/R_0$. It is often more convenient to use the damping factor (or decrement) $d = Q^{-1}$. The decrement of the circuit without the sample is, therefore, $d_0 = R_0/ωL_0$. On inserting the sample into the coil the circuit decrement changes to $d = (R_0 + kR_c)/[ω(L_0 + kL_c)]$. In the case that the sample causes merely small changes in the circuit inductance ($kL_c ≪ L_0$) and the circuit quality without the sample is fairly high ($ωL_0/R_0 ≫ 1$), the expression for the difference $d - d_0 = PA$ can be written as

$$PA = kR_c/ωL_0 = kR_c/R_0Q_0.$$  

(3)

It is seen from it that $PA$ is determined by the loss in the sample. For the further analysis of the situation it is useful to consider the following approximations: $tan(δ_ε) ≫ 1$ and $tan(δ_μ) ≪ 1$. This enables us to write the Eq. (3) in the form

$$PA = k_0 \left(1 + \frac{1}{2 ρ_{sp}} \right) R_s$$  

(4)

where $k_0 = k/(ωL_0)$, $R_s = (ωμ'/2σ_{ef})^{1/2} = (ωμρ_{rf}/2)^{1/2}$ is the surface resistance, $ρ_{rf}$ is the RF resistivity. The Eq. (4) present a simplified way to take into account the influence of the conductivity and permeability on the RF loss.

III. RESULTS AND DISCUSSION

The temperature dependence of the real part, $μ_r$, of the relative AC permeability of the sample studied is shown in Fig. 1. The value of $T_C ≈ 250$ K is found if $T_C$ is defined as the temperature of the inflection point in the $μ_r(T)$ curve. In general, the behavior of $μ_r(T)$ agrees well with the known studies of the AC susceptibility in ceramic La$_{0.5}$Sr$_{0.5}$CoO$_{3-δ}$ [11, 12]. When going from high to lower temperature, the relative AC permeability increases slightly between 320 K and 250 K, then it...
FIG. 1: Temperature dependences of the real part of the relative magnetic permeability, $\mu_r = \mu / \mu_0$, of the sample La$_{0.5}$Sr$_{0.5}$CoO$_3$ in AC magnetic field $H_{ac} = 4$ Oe and frequency 10 kHz. The dependences have been recorded in zero field (open circles) and in DC field 40 Oe (solid circles) with temperature increasing after the sample was cooled in zero field.

increases sharply at $T = T_c \approx 250$ K and reaches its maximum at a temperature $T_{\text{max}} = 232$ K. A considerable decrease in $\mu_r(T)$ (or AC susceptibility) below $T_{\text{max}}$ (Fig. 1) is a well-known feature of cobaltites. It is attributed to the cluster-glass behavior [2] or to an increase in the coercivity with decreasing temperature [12]. Figure 1 demonstrates a high sensitivity of the $\mu_r(T)$ to an external magnetic field: even a fairly low field (40 Oe) causes a considerable decrease in $\mu_r(T)$. The effect is maximal at $T = T_{\text{max}}$. When going to either side from $T_{\text{max}}$, the influence of the magnetic field becomes weaker. In particular, it is negligible for $T \leq 100$ K.

The temperature dependence of the DC resistivity, $\rho(T)$, is shown in Fig. 2. The $\rho$-values were calculated taking into account the porosity of the sample [10]. The $\rho(T)$ behavior is metallic ($d\rho/dT > 0$) over the whole temperature range investigated, below and above $T_c$. This behavior (and the resistivity values) agree with the known data on ceramic La$_{0.5}$Sr$_{0.5}$CoO$_3$ [2]. The $\rho(T)$ dependence exhibits a change of slope at $T_c$ (see insert in Fig. 2) due to a contribution from the electron scattering on the spin disorder (in addition to the usual contributions from crystal lattice defects and electron-phonon scattering) [13]. This “magnetic” contribution to the resistivity, $\Delta \rho_m$, depends on the magnetization. The external magnetic field enhances the spin order (that is, the magnetization), which leads to a decrease in the resistivity, that is to negative MR. The magnetic-field dependences of resistance of the sample studied are shown in Fig. 3. It is seen that the MR depends strongly on temperature. The temperature dependence of the MR at $H = 20$ kOe is presented in Fig. 4. This behavior is quite common for FM oxides (like manganites and
The MR is maximal at \( T \approx T_c \). It goes down rather steeply for temperature deviating to either side from \( T_c \). This temperature behavior of the MR is expected for FM oxides of fairly good crystal perfection or for polycrystalline samples with rather strong connectivity between the crystallites. Indeed, the MR amplitude is determined by the ability of an external magnetic field to increase the magnetization. It is obvious that at low temperatures \( (T \ll T_c) \), when nearly all spins are already aligned by the exchange interaction, this ability is minimal. For increasing temperature and, especially, at temperature close to \( T_c \), the magnetic order becomes weaker (the magnetization goes down) due to thermal fluctuations. In this case the possibility to strengthen the magnetic order with an external magnetic field increases profoundly. This is the reason for maximal MR magnitude near \( T_c \). Above \( T_c \), the spin arrangement becomes essentially random, the magnetization is zero, and, therefore, the MR is close to zero as well.

Consider now the behavior of the RF loss \( P_A \) in the sample studied. We begin with the temperature behavior \( P_A(T) \) in zero field (Fig. 4). In doing so we will refer to Eq. 4. It is worthwhile to mention here that certain mutual correlations in the temperature behavior of \( \mu \) and \( \mu^\prime \) can be expected. It follows immediately from the Kramers-Kronig relations. These are helpful in consideration of a remarkable correlation between the temperature behavior of the loss and the permeability \( \mu_r \) found in this study. The correlation is evident at once on comparing Figs. 4 and 5. It is clear that far enough above \( T_c \) the relations, \( \mu_r = 1 \) and \( \mu^\prime = 0 \), are true. In this case, according to Eq. 4 the loss depends only on the conductivity. In FM oxides, like mixed-valence manganites and cobaltites, the FM fluctuations (or developing of small FM regions in the paramagnetic matrix) can be evident, however, fairly far above \( T_c \) for extrinsic and intrinsic sources of magnetic inhomogeneity. In that event, as the \( T_c \) is approached from above, the slight increase in the susceptibility or permeability can be observed even in the range far from \( T_c \). This will cause a corresponding increase in the loss. The drastic increase in \( P_A \) occurs, however, only quite near \( T_c \) due the sharp increase in \( \mu^\prime \) at transition of the most of the sample volume to the FM state (Figs. 4 and 5). Notice that the conductivity increases with temperature decreasing in the whole temperature range studied (Fig. 2) and this (according to the Eq. 4) favors a decrease in the loss. Contribution of the permeability to temperature variations of the loss is, however, far more than that of the conductivity, so, as a result, the loss increases at the transition to the FM state. For further decrease in temperature below \( T_c \) the loss comes to some maximal value at \( T \approx 230 \text{ K} \) and, after that, goes down (Fig. 5). All this correlates well with the \( \mu_r(T) \) behavior (Fig. 5). Even the temperatures of the maxima for \( P_A \) and \( \mu_r(T) \) curves are both at about 230 K.

It is seen that the RF loss is far more sensitive to strengthening of the spin order at the transition to the FM state than the DC conductivity. Really, \( \rho(T) \) shows only a change in the slope at \( T_c \) that means the steeper decrease in resistivity with temperature decreasing below \( T < T_c \) (Fig. 5). In contrast to this, the RF loss increases sharply (nearly twice) in the vicinity of \( T_c \) in zero field (Fig. 5). This indicates that it is precisely the changes
in the permeability that cause the large variations in the loss near $T_c$.

From measurements of the loss, $P_A$, at different magnetic fields we have obtained the values of the magnetoabsorption (MA), which has been defined as $[P_A(H) - P_A(0)]/P_A(0) = \Delta P_A(H)/P_A(0)$. Using the abovementioned approximation, $\tan(\delta_{\mu}) = \mu''/\mu' \ll 1$, and taking into account Eq. 4, a simplified expression for MA can be written as

$$\Delta P_A(H)/P_A(0) \approx \frac{R_s(H)}{R_s(0)} - 1 = \sqrt{\frac{\mu'(H)\rho_{rf}(H)}{\mu'(0)\rho_{rf}(0)}} - 1.$$

This shows immediately the correlation between the $P_A(T)$ and $\mu'(T)$. It is clear from Eqs. 4 and 5 that owing to decrease in $\rho_{rf}$ and $\mu'$ in an applied magnetic field, the MA should be negative. This is consistent with the results found. We actually found a high sensitivity of the RF loss to applied magnetic field (Figs. 6, 7, 8). It is remarkable that the strong magnetoabsorption effect shows itself in rather low magnetic fields. The effect is really giant when compared with the DC magnetoresistance. Not far from $T_c$, at $T_{\text{max}} \approx 230$ K, the MA is about 38% in field $H = 2.1$ kOe (Fig. 8). By contrast, the MR is about 0.26% near $T_c$ in the above-mentioned field (Fig. 8) and it has increased no more than to about 2.15% at field $H = 20$ kOe (Figs. 8 and 9).

The reason why the magnetoabsorption effect is far stronger than the MR one appears rather obvious. As indicated above, the MR in the vicinity and below $T_c$ is determined by the enhancing of the spin order (magnetization) in an applied magnetic field. This leads to the conductivity increasing (that is to the negative MR).

Unlike the DC magnetoresistance, the magnetoabsorption is determined not only by influence of the magnetic field on the conductivity, but, to a much greater extent, by changes in the permeability in the magnetic field. The remarkable correlation between behaviors of the $P_A(T)$ and the $\mu'(T)$ was mentioned above. No less noteworthy correlation can be found between the behaviors of $P_A$ and the $\mu'$ in an applied magnetic field (compare Figs. 7 and 8).

It follows that the giant magnetoabsorption effect is qualitatively explicable taking into account a decrease in the DC permeability. The RF resistivity, $\rho_{rf}$, decreases in magnetic field as well and, therefore, gives also some contribution to this effect. It could be expected [10], that RF resistivity is more sensitive to magnetic field than DC resistivity. We cannot, however, separate a contribution of RF resistivity to the magnetoabsorption. In any case, however, the above-described results testify that this contribution is negligible as compared to that of the permeability.

No direct measurements of $\mu''$ in the La$_{0.5}$Sr$_{0.5}$CoO$_{3-\delta}$ or other Sr-doped cobaltites have been made, to our knowledge. It is known that the value of $\mu'$ decreases with increasing frequency [13], but it can be large for fairly low frequencies. This tendency, general for FM metals, is found to hold for related perovskite-like oxides as well. For example, for polycrystalline manganite La$_{0.3}$Sr$_{0.7}$MnO$_3$ the magnitude of $\mu'$ is fairly close to unity at $\nu = 100$ MHz, but in the 1-MHz-range it can be significantly larger than unity [13]. In this case the relation, $\mu''/\mu' \ll 1$, holds.

In conclusion, we found the giant negative RF magne-
to absorption effect in bulk sample of La$_{0.5}$Sr$_{0.5}$CoO$_{3-\delta}$ under low DC magnetic fields. The effect should be attributed primarily to the changes in the magnetic permeability in applied magnetic field. Such low-field magnetoabsorption may be of potential application in RF devices controlled by magnetic field [16]. It is clear that effective temperature range for application of any material with giant magnetoabsorption is determined by its $T_c$ value. For the range near or above room temperature, the manganite La$_{1-x}$Sr$_x$MnO$_3$ with 0.25 < $x$ < 0.35 is suggested.

[1] M. Itoh, I. Natori, S. Kubota, and K. Motoya, J. Phys. Soc. Jap. 63, 1486 (1994).
[2] P. Ganguly, P. S. Anil Kumar, P. N. Santosh and I. S. Mulla, J. Phys.: Condens Matter 6, 533 (1994).
[3] M. A. Señarís-Rodríguez and J. B. Goodenough, J. Sol. State Chem. 118, 323 (1995).
[4] R. Mahendiran and A. K. Raychaudhuri, Phys. Rev. B 54, 16044 (1996).
[5] V. Golovanov, L. Mihaly, and A. R. Moodenbaugh, Phys. Rev. B 53, 8207 (1996).
[6] J. M. D. Coey, M. Viret, and S. von Molnar, Adv. Phys. 48, 167 (1999).
[7] E. Dagotto, T. Hotta, and A. Moreo, Phys. Rep. 344, 1 (2001).
[8] A. B. Beznosov, B. I. Belevtsev, E. L. Fertman, V. A. Desmenko, D. G. Naugle, K. D. D. Rathnayaka and A. Parasiris, Fiz. Nizk. Temp., 28, 774 (2002) [Low Temp. Phys., 28, 556 (2002)].
[9] A. B. Beznosov, P. P. Pal-Val, B. I. Belevtsev, V. B. Krasovitsky, E. L. Fertman, L. N. Pal-Val, I. N. Chukanova and T. G. Deyneka, Izv. AN, Ser. Fiz., 66, 758 (2002).
[10] B. I. Belevtsev, N. T. Cherpak, I. N. Chukanova, A. I. Gubin, V. B. Krasovitsky, and A. A. Lavrinovich, J. Phys.: Condens. Matter 14, 2591 (2002).
[11] S. Mukherjee, R. Ranganathan, P. S. Anilkumar, and P. A. Joy, Phys. Rev. B 54, 9267 (1996).
[12] P. S. Anil Kumar, P. A. Joy, and S. K. Date, J. Phys.: Condens. Matter 48, 167 (1999).
[13] S. V. Vonsovsky, Magnetism, (Nauka, Moscow, 1971).
[14] R. M. White, Quantum theory of magnetism, (Springer-Verlag, New York, 1983).
[15] Jinhui Wang, Gang Ni, Wenli Gao, Benxi Gu, Xiabin Chen, and Youwei Du, phys. stat. sol. (a) 183, 421 (2001).
[16] M. Knobel and K. R. Pirota, J. Magn. Magn. Mater. 242-245, 33 (2002).