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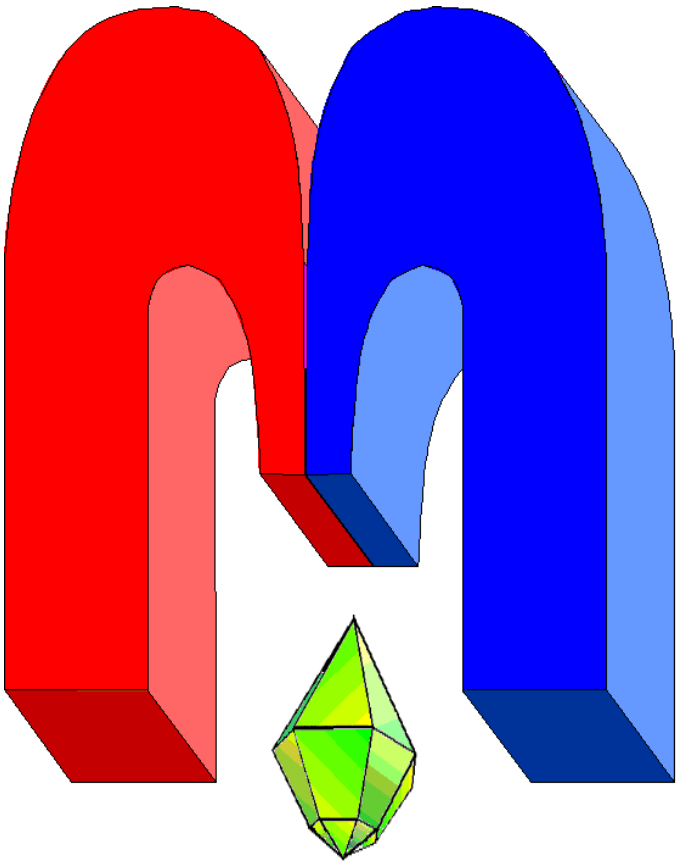

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* In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

Electron spin resonance study of the demagnetization fields of the ferromagnetic and paramagnetic films†

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The results of the electron spin resonance study of the $La_{1-x}Ca_xMnO_3$ manganite and the diphenyl-picrylhydrazyl thin films for the magnetic field parallel and perpendicular to plane of the films are presented. The temperature dependence of the demagnetizing field is obtained. The parameters of the Curie-Weiss law are estimated for the paramagnetic thin film.

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1. Introduction

It is well know that the electron spin resonance (ESR) is convenient tool for studying the local magnetic fields. One of the source of local magnetic fields is a demagnetizing field. It is due to the presence of magnetic poles of ferromagnetic body. The demagnetizing field influence on the ferromagnetic resonance (FMR) spectrum is well described by C. Kittel [1]. His equations connect the FMR signal position with the demagnetizing vector components and agree well with results of FMR study of magnetic samples with different shape and size (see for example [2,3]). These equations have a simple form for a thin film ($d \ll l$, where d is the film thickness and l is the lateral size) oriented parallel or perpendicular to the applied magnetic field direction. Applying the Kittel theory to the FMR study results on can estimate the saturation magnetization M_s value. In present work this will be demonstrated with the example of the $La_{1-x}Ca_xMnO_3$ manganite thin film. We will also show that the demagnetizing field effect can be find in a weak magnetized (paramagnetic) film. Thereby one can obtain the susceptibility value estimation. We will demonstrate this with the example of the ESR study of the diphenyl-picrylhydrazyl (DPPH) thin film.

In order to analyze experimental results we will present the Kittel equations.

2. Kittel equations

The main result of the Kittel work [1] can be written in the following equation:

$$H_{\mathrm{R0}}^2 = \left(H_{\mathrm{a}} + \left(N_y - N_z\right) M\right)\left(H_{\mathrm{a}} + \left(N_x - N_z\right) M\right), \tag{1}$$

$$H_{\mathrm{R0}} = \frac{h\nu}{g \cdot \mu_{\mathrm{B}}}. \tag{2}$$

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Here H_{R0} is the resonance field, H_a is the applied (external) field, where the resonance signal is observed, M is the magnetization, N_x, N_y, N_z are the demagnetizing vector components.

If some components become equaled to zero, this equation takes a simpler form. For example, in the case of the film perpendicular to the applied field direction $N_x = N_y = 0$, $N_z = 4\pi$ (CGS units), and then the equation gets the following form:

$$H_{R0} = H_a - 4\pi M. \tag{3}$$

It means that the resonance field is the sum of the applied field and the demagnetizing one. For parallel orientation ($N_x = N_z = 0$, $N_y = 4\pi$) the equation (1) becomes as follows:

$$H_{R0} = \sqrt{H_a\,(H_a + 4\pi M)}. \tag{4}$$

It is easy to apply these equations for analyzing the FMR spectra.

3. FMR results of the $La_{1-x}Ca_xMnO_3$ thin film

We use the thin film of manganite $La_{1-x}Ca_xMnO_3$ as a ferromagnetic sample. The epitaxial single-crystalline film of 10 nm thickness was deposited on the MgO substrate. We chose the sample with the Ca concentration $x = 0.4$, which becomes ferromagnetic below $T_c = 250$ K.

ESR spectra were recorded by ESR spectrometer Bruker BER-418s with frequency of about 9.2 GHz. The temperature was changed by the helium flow cryostat.

Fig. 1 demonstrates the ESR spectrum variation with temperature in the two orientations of the film plane with respect to the field direction. Every spectrum contains of one broad line from the Mn ions of the $La_{0.6}Ca_{0.4}MnO_3$ compound and the narrow signal set near $g \approx 2$ which are due to impurities in the MgO substrate.

At high temperatures the broad line and the narrow signals are superimposed. Upon decreasing the temperature the ferromagnetic ordering of the Mn ions occurs and the broad signal shifts to the lower fields at the parallel orientation and to the higher fields at the perpendicular orientation.

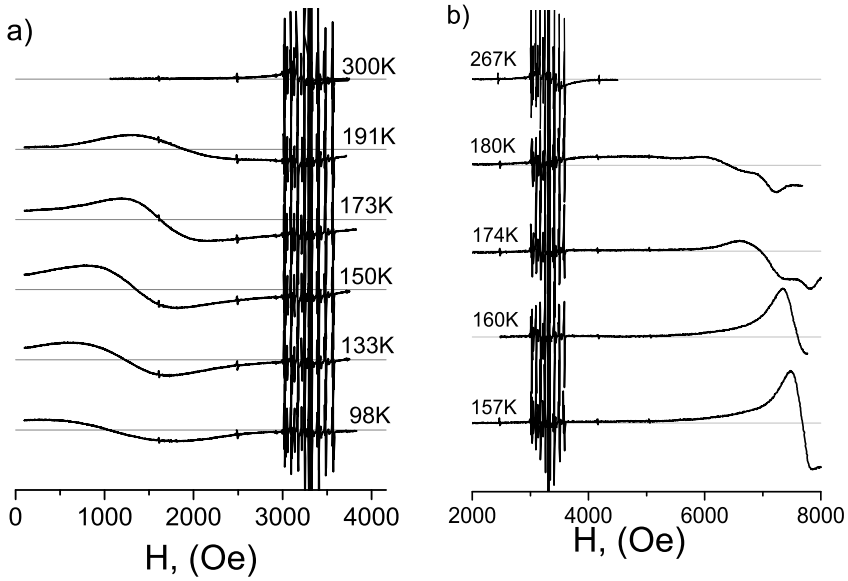

Figure 1. ESR spectra of the $La_{0.6}Ca_{0.4}MnO_3$ film on the MgO substrate for parallel (a) and perpendicular (b) orientations of the sample in the magnetic field at different temperatures.

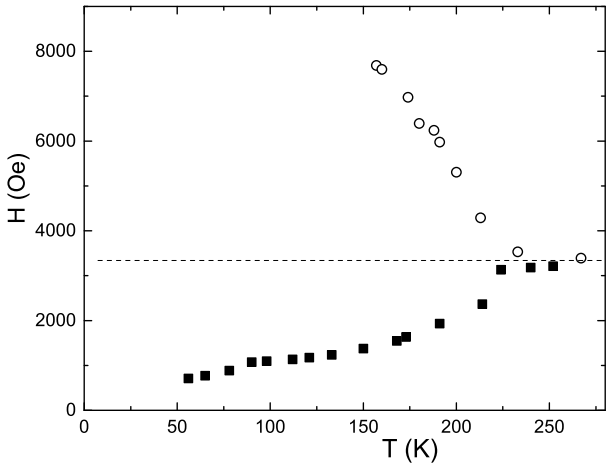

Figure 2. Temperature dependences of the Mn resonance fields for parallel (squares) and perpendicular (circles) orientations of the film.

The temperature dependence of the Mn resonance filed obtained from the spectra is shown in Fig. 2 for the both orientations. In perpendicular orientation the signal shifts fast and goes beyond the available field range at 150 K.

For describing the date obtained at low temperature, when the sample is in ferromagnetic state, we can use the Kittel equations (3) and (4), and estimate the magnetization magnitude.

One can assume that H_{R0} is equal to the broad signal resonance field in the paramagnetic state, where the magnetization is small. Estimations of the magnetization ($4\pi M$) give 4793 Oe for parallel orientation and 3105 Oe for perpendicular orientation at 180 K.

For perpendicular orientation the demagnetizing field equals $4\pi M$. When the applied field is parallel to the film plane the demagnetizing field contribution is considerably smaller then $4\pi M$ as one can see from Fig. 2 and equation (4).

4. ESR results of the DPPH film

The magnetization of paramagnetic sample is considerably smaller (in several orders of magnitude) than magnetization of ferromagnetic one. So the demagnetizing field effect can be found if the ESR signal width is not too larger than the demagnetizing contribution. The DPPH signal has this property. Its width is near 1.5 Oe. Therefore we use the DPPH thin film to estimate the demagnetization effect.

In order to detect a weak shift of the DPPH signal we employ the signal of lithium as a reference signal. The lithium signal has a narrow width ($\delta H = 0.1$ Oe) and well-defined g-factor ($g(\mathrm{Li}) = 2.00226$ [4]).

The spectra of the DPPH film and the lithium dendrites were recorded in the two orientations of the field with respect to the film plane (parallel and perpendicular) in the temperature range from 20 K to 300 K with the step $5 \div 20$ K. Some spectra is shown in Fig. 3. As one can see, the lithium signal position does not change with temperature, and the DPPH signal is shifted. We determine the DPPH signal position with respect to the lithium signal.

One can analyze the shift using Kittel equations. They allow one to estimate the magnetization value. It is directly proportional to the magnetic susceptibility. The susceptibility of paramagnetic sample changes with temperature according to the Curie-Weiss law. This determines the DPPH signal shift. Thus we can obtain the dependence of the demagnetization field on a temperature.

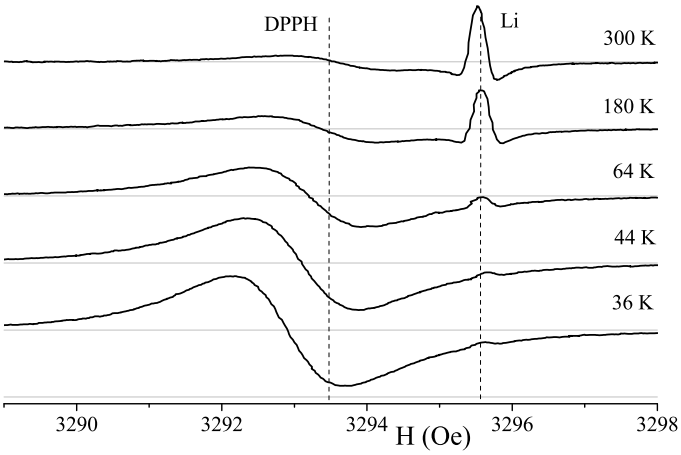

Figure 3. ESR spectra of DPPH film for parallel orientation at different temperatures.

In order to obtain the demagnetizing field from the ESR spectrum it is necessary to define the resonance fields of DPPH and lithium. The resonance field of lithium is defined by its g-factor:

$$H_{\mathrm{Li}} = \frac{h\nu}{g_{\mathrm{Li}} \cdot \mu_{\mathrm{B}}}. \tag{5}$$

The DPPH signal position is determined not only by g-factor, the demagnetizing field contributes as well:

$$H_{\mathrm{DPPH}} = \frac{h\nu}{g_{\mathrm{DPPH}} \cdot \mu_{\mathrm{B}}} + \Delta H_{\mathrm{df}}. \tag{6}$$

Hence, the distance between the signals is expressed as follows:

$$\Delta H = \frac{h\nu}{g_{\mathrm{Li}} \cdot \mu_{\mathrm{B}}} - \frac{h\nu}{g_{\mathrm{DPPH}} \cdot \mu_{\mathrm{B}}} - \Delta H_{\mathrm{df}}. \tag{7}$$

We can obtain the demagnetizing field contribution if we remove contribution defined by g-factors:

$$\Delta H_{\mathrm{df}} = \Delta H - \left(\frac{h\nu}{g_{\mathrm{Li}} \cdot \mu_{\mathrm{B}}} - \frac{h\nu}{g_{\mathrm{DPPH}} \cdot \mu_{B}} \right). \tag{8}$$

This procedure can be performed for all spectra obtained at different temperatures. As a result, we get the temperature dependence of the demagnetizing field contribution of DPPH film. This dependence is shown in Fig. 4 for parallel orientation (a) and for perpendicular orientation (b).

From these data we can extract the DPPH susceptibility value and its temperature dependence. To do this one use Kittel equations (3) and (4) with taking into account the relation $M = \chi H_{\mathrm{a}}$. Then for perpendicular orientation the equation (3) becomes as follows:

$$H_{\mathrm{R0}} = H_{\mathrm{a}} \left(1 - 4\pi\chi\right), \tag{9}$$

where χ is the DPPH susceptibility. In the case of parallel orientation the equation has the form:

$$H_{\mathrm{R0}} = H_{\mathrm{a}} \sqrt{1 + 4\pi\chi}. \tag{10}$$

In paramagnetic phase the susceptibility temperature dependence obeys Curie-Weiss law:

$$\chi = \frac{C_{\chi}}{T - \theta}. \tag{11}$$

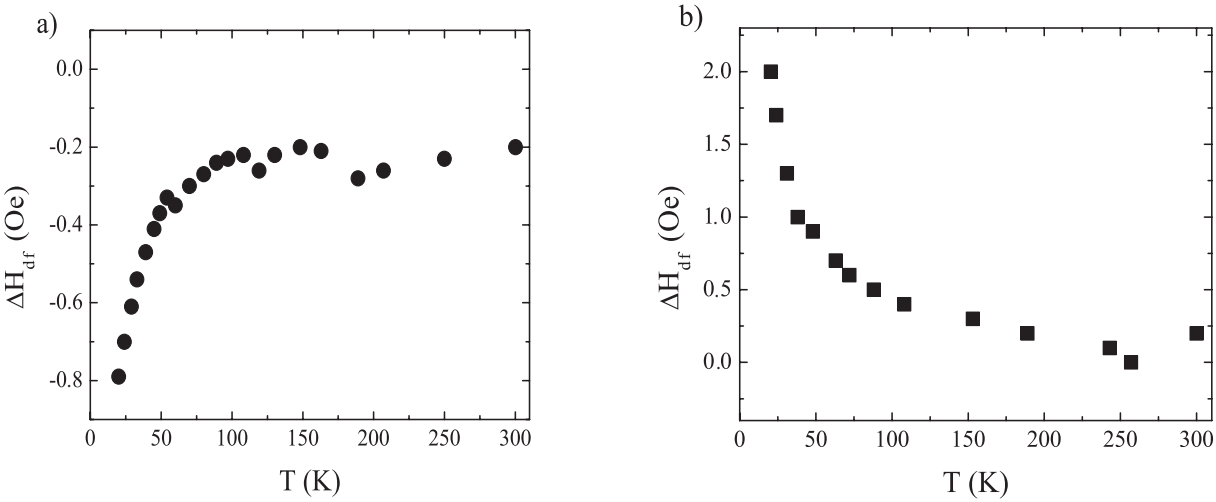

Figure 4. Temperature dependence of the demagnetizing field contribution for the magnetic field parallel (a) and perpendicular (b) to plane of the DPPH film.

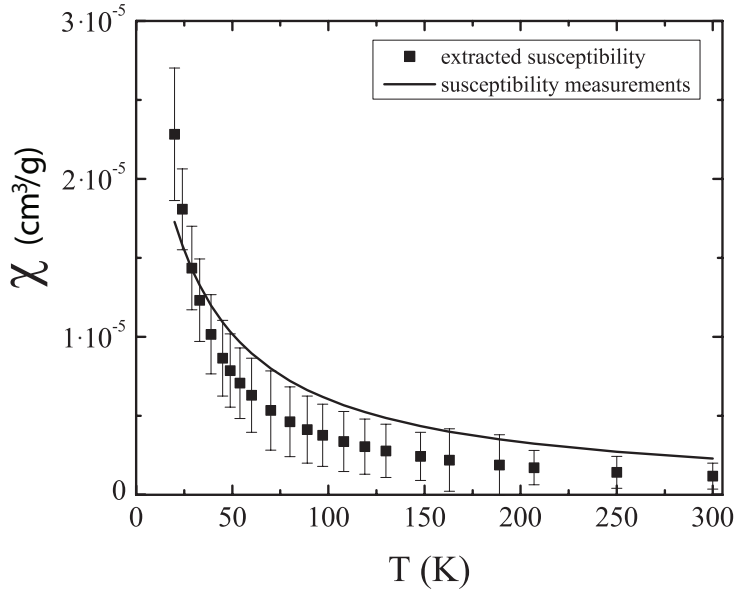

Figure 5. Temperature dependence of the DPPH susceptibility. Squares show the data obtained by ESR measurements. The solid line is data of the direct susceptibility measurements [5].

By substituting equation (11) in Kittel formulas (9) and (10) we can obtain expressions for the field, where the resonance signal is observed in perpendicular and parallel orientations:

$$H_{\mathrm{a}} = \frac{H_{\mathrm{R0}}}{1 - 4\pi\frac{C_\chi}{T-\theta}}, \tag{12}$$

$$H_{\mathrm{a}} = \frac{H_{\mathrm{R0}}}{\sqrt{1 + 4\pi\frac{C_\chi}{T-\theta}}}. \tag{13}$$

These equations describe temperature dependence of resonance field and give opportunity to estimate the parameters of the Curie-Weiss law: $\theta \approx 20\,\mathrm{K}$, $C_\chi = 6 \cdot 10^{-4}$ K·cm^3/g. The dependence (11) with these parameters is plotted in Fig. 5 along with results of the direct susceptibility measurement [5]. It is seen they are in well agreement.

Thus, we have demonstrated the demagnetizing field effect on the ESR spectra of the thin ferromagnetic and paramagnetic films. Even for paramagnetic sample this effect is well observable and allow us to obtain useful information.

Acknowledgments

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