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Magnetic Pseudoresonance in Manganite Thin Films

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Abstract A strong narrow peak in the field dependence of the radio-frequency absorption (the magnetic pseudoresonance) has been found and investigated in epitaxial thin films of $La_{2/3}Sr_{1/3}MnO_3$ possessing uniaxial magnetic anisotropy in the film plane. The peak is observed when the in-plane external magnetic field **H** is directed perpendicular to the easy axis and equals to the anisotropy field H_u. The frequency dependence of the peak magnitude measured in the frequency range of 10–300 MHz approximately follows the Debye law behavior with the characteristic relaxation time of about 2.2 ns. The physical model of the phenomenon is suggested, based on the giant increase in static transversal susceptibility due to a sharp reorientation of the equilibrium magnetization when approaching the pseudoresonance conditions.

1 Introduction

Among numerous trends of current thin-film electronics, a significant place is occupied by structures and devices employing unique magnetic and transport properties of doped rare-earth manganites $Ln_{1-x}A_xMnO_3$, where A is a bivalent alkaline-earth element. In recent years, these materials attracted considerable

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interest owing to the effect of colossal magnetoresistance and rich phase diagram, including the combined magnetic and metal–insulator phase transitions (see for example the review article [1]). Possible applications of the manganite thin films and nano-structures as elements of spintronics were discussed in Refs. [2–4]; it was found that one of central problems is the magnetic anisotropy arisen during the film synthesis. In particular, when growing the epitaxial films of $La_{2/3}Sr_{1/3}MnO_3$ (LSMO), an additional in-plane uniaxial magnetic anisotropy is created due to mismatch between the film and substrate [5, 6]. Under some conditions, this axial anisotropy (especially important for applications) may strongly exceed the natural cubic-symmetry magnetic anisotropy of LSMO. The uniaxial anisotropy energy K_u and the direction of the easy axis $\mathbf{n_u}$ depend on the material and crystallographic orientation of the substrate, as well as on the synthesis conditions and temperature [6–8]. At room temperature, the value of the 1st order anisotropy field $H_u = 2K_u/M_0$, where M_0 is the saturated equilibrium magnetization, can amount hundreds Oe.

The magnetic anisotropy can be studied with the technique of ferromagnetic resonance (FMR). The FMR spectra contain comprehensive information on the magnitude, symmetry and direction of the internal fields. This method was used by the authors of Ref. [4–6], where the data obtained on the standard EPR spectrometer working in the microwave range (at the frequency of $\sim$9.5 GHz) were supplemented by measurements in the radio-frequency (r.f.) range ($\sim$300 MHz) performed with the original home-made setup [9]. In the course of the work, an intense narrow peak of r.f. absorption was observed at the in-plane orientation of the external magnetic field $\mathbf{H} \perp \mathbf{n_u}$ when sweeping through the value of $H = H_u$. The discovered signal resembled a line of magnetic resonance; however, its field position and shape were found to be independent on frequency in a wide range. Thus, the peak-shape of the "resonance" feature is only revealed in the dependences of the absorption on the magnetic field magnitude and direction. So we will call this phenomenon "pseudoresonance". Note that a similar effect was observed previously in ferromagnetic metal alloys [10], and some theoretical description was suggested [10–12]. The present work is aimed to a more detailed experimental study of the pseudoresonance phenomenon in epitaxial LSMO films, together with advancing an alternative approach to the theoretical model. Besides, the prospects of using this effect in studying magnetic anisotropy, spin dynamics and relaxation will be discussed.

2 Experiment

The $La_{2/3}Sr_{1/3}MnO_3$ epitaxial films of 50 nm thick were grown by laser ablation on the single-crystal substrates of $NdGaO_3$ [5]. The geometry of the experiment is illustrated schematically in Fig. 1. The sample of size 5×5 mm^2 cut with its edge parallel to the direction of $\mathbf{n_u}$ was placed in the central area of the pick-up coil of the resonant r.f. Q-meter. The film could be oriented by rotation around its normal with the high-precision goniometer in such a way that both the magnetic r.f. field $\mathbf{h}$ (vertical) and the external static field $\mathbf{H}$ (horizontal) remained in the film plane. In the following, the angle between $\mathbf{n_u}$ and $\mathbf{H}$ is denoted by θ_H, whereas θ_0 is the angle

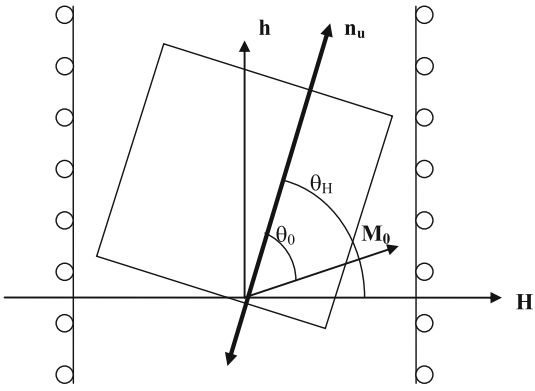

Fig. 1 Sketch of the experimental geometry. The *square* represents the film inside the r.f. coil, the sample edge being parallel to the easy axis $\mathbf{n_u}$. Directions of the static and r.f. fields, as well as the sample magnetization are indicated by the *arrows*

between the equilibrium sample magnetization $\mathbf{M_0}$ (also lying in the film plane) and $\mathbf{n_u}$.

The home-built spectrometer operating at the frequency of $\omega/2\pi \cong 300$ MHz [9], as well as a series of Q-meters covering the range of 10–40 MHz were used to measure the imaginary part of dynamic magnetic susceptibility $\chi''(\omega)$, which corresponds to the r.f. absorption coefficient. The absolute values of χ'' were calibrated using EPR absorption of the DPPH free radical placed into the same coil as a reference; the corresponding resonance fields fell in the range of 25–110 Oe. All measurements were carried out at the room temperature.

A typical signal of the pseudoresonance absorption recorded at 300 MHz and $\theta_H = \pi/2$ is shown in Fig. 2a. The maximum of the absorption is observed at $H = (226 \pm 1)$ Oe, in excellent agreement with the value of H_u as determined from the angular dependence of the FMR spectrum. The width of the "resonance" peak at the half height amounts to 11 Oe, which can be compared with the FMR line width $\delta H_{FMR} = 19$ Oe as measured in the same sample at 9.5 GHz. Note that the peak absorption exceeds the value of $\chi_0 \equiv M_0/H$ by more than an order of magnitude [in this sample $M_0 = (340 \pm 10)$ Oe].

The angular dependence of the pseudoresonance peak magnitude is demonstrated in Fig. 2b. It is seen that a narrow "resonance" occurs in this case as well, with the center at $\theta_H = \pi/2$ and the width of a few tenth degree. Hence, the magnetic pseudoresonance signal can be found and recorded only at accurate adjusting to the pseudoresonance conditions ($\theta_H = \pi/2$; $H = H_u$).

The frequency dependence of the peak magnitude of χ'' is presented in Fig. 3. In the whole frequency range studied, the measured values of χ'' were found to be independent on the amplitude h of the r.f. field which was varied in the order of magnitude range up to $\sim 2 \times 10^{-3}$ Oe. As seen from Fig. 3, the absorption coefficient initially rises approximately linearly with ω increasing, but then bends down, evidently passing through a broad maximum somewhere near 100 MHz. Such a frequency dependence is typical of the absorption caused by relaxation processes and can be exploited to determine the relaxation rate. We will discuss this opportunity in the next section.

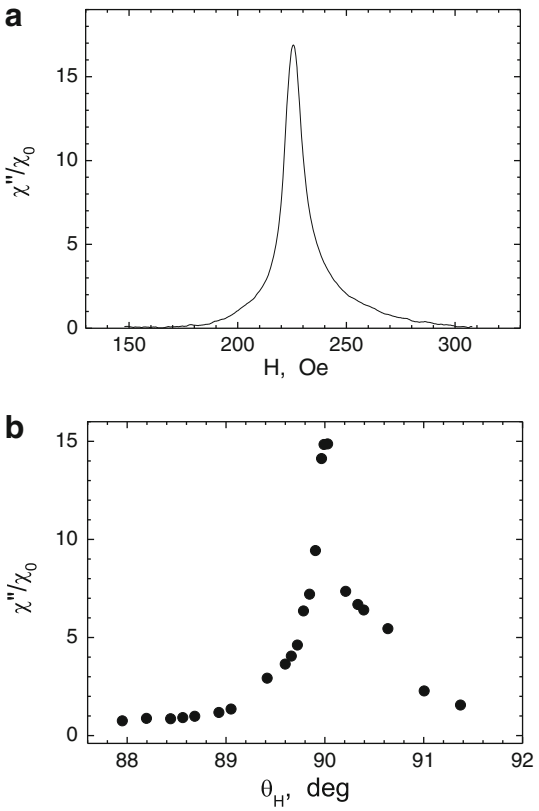

Fig. 2 Imaginary part of dynamic susceptibility χ'' measured at $\omega/2\pi = 300$ MHz in the vicinity of the specific point. **a** In dependence on the field H at $\theta_H = \pi/2$; **b** in dependence on the angle θ_H at $H = H_u$. The χ'' values are normalized to $\chi_0 \equiv M_0/H_u$

3 Discussion

Let us discuss the physical origin of the magnetic pseudoresonance by using a conventional procedure of evaluating the dynamic magnetic susceptibility of a ferromagnetic film with account made for the demagnetization tensor and uniaxial magnetic anisotropy with the easy axis $\mathbf{n_u}$. To this end, the Landau-Lifshits-Gilbert (LLG) equation is commonly used [13]:

$$\partial\mathbf{M}/\partial t = -\gamma\mathbf{M} \times \mathbf{H_i} + (\alpha/M)\mathbf{M} \times \partial\mathbf{M}/\partial t, \quad (1)$$

where $\mathbf{H_i}$ is the effective (internal) magnetic field including both the static and r.f. components and accounting for demagnetization and magnetic anisotropy and α is the damping parameter. We begin with looking for the field dependence of the FMR frequency, $\omega_0(H)$. For this purpose, one can use Eq. 1 in the mode of free oscillations ($h = 0$) without damping ($\alpha = 0$):

$$\partial\mathbf{M}/\partial t = -\gamma\mathbf{M} \times \mathbf{H_{i0}}, \quad (2)$$

where $\mathbf{H_{i0}}$ is the static component of $\mathbf{H_i}$. Simultaneously, the condition of thermodynamic equilibrium must be fulfilled. Supposing $M_0 = \text{const}$, one can determine the angle θ_0 between the equilibrium direction of $\mathbf{M_0}$ and $\mathbf{n_u}$. Standard

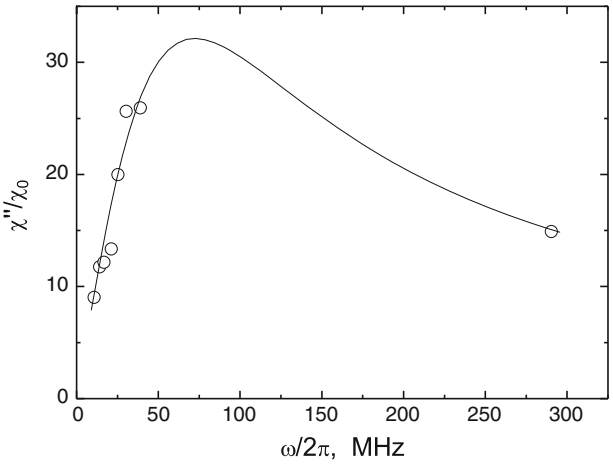

Fig. 3 Peak value of χ'' under the pseudoresonance conditions as a function on frequency. The *curve* corresponds to the Debye law, Eq. 7

approach based on minimization of the free energy, including the Zeeman interaction with the field **H**, uniaxial magnetic anisotropy of the 1st order, and demagnetizing field leads to the equation:

$$\sin(2\theta_0) = (2H/H_u)\sin(\theta_H - \theta_0) \tag{3}$$

(Note that this equation coincides with that obtained for a ferromagnetic sphere with axial anisotropy [13].) Limiting oneself by linear approximation in Eq. 2 and setting the determinant of combined equations for the oscillating magnetization components to zero, one gets for $\theta_H = \pi/2$:

$$\omega_0^2 = \begin{cases} \gamma^2 H_u(H_u + 4\pi M_0)\left[1 - \left(\frac{H}{H_u}\right)^2\right], & H \le H_u \\ \gamma^2(H + 4\pi M_0)(H - H_u), & H > H_u \end{cases} \tag{4}$$

The plot of $\omega_0(H)$ is shown in Fig. 4. It is seen that the FMR frequency equals zero at the combination of two conditions, $\theta_H = \pi/2$ and $H = H_u$, thus forming a specific point in the $\omega_0(H)$ dependence.

Evaluation of the magnetic susceptibility in this specific point is not trivial. In particular, linearization of the full LLG Eq. 1 is hardly executable at $\omega_0 \to 0$. In Refs. [10–12], this problem was overcome by numerical calculations of the magnetization evolution in non-linear regime, including sophisticated trajectories between two potential wells on each side of the right angle. This leads, however, to a non-linear dependence of the r.f. absorption on the amplitude of the radio frequency field, h, in contradiction with our data. That is why we use a different approach to analyze the linear response (susceptibility) in this specific point. Namely, we divide the problem into two independent stages: (1) evaluation of static susceptibility $\chi(0) \equiv \chi^0$; and (2) calculation of the frequency dependence $\chi(\omega)$.

The first problem can be solved by using the above mentioned condition of thermodynamic equilibrium, Eq. 3. The solution shows that the anisotropy field tries

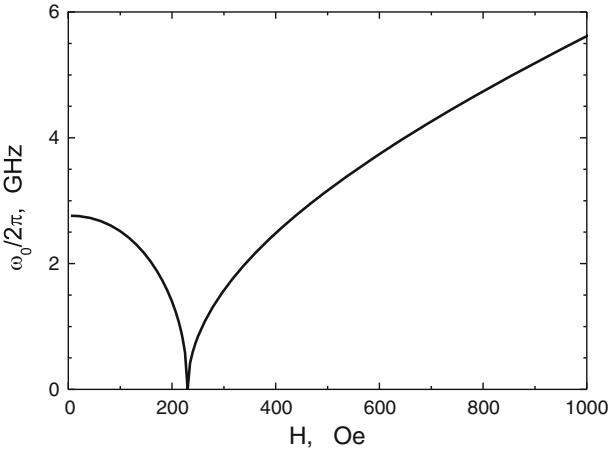

Fig. 4 FMR frequency as a function of the external magnetic field at $\theta_H = \pi/2$. The plot is calculated according to Eq. 4 with the parameters of the sample under study

to keep the $\mathbf{M_0}$ vector close to the easy axis. As a result, $\mathbf{M_0}$ deviates strongly from the direction of $\mathbf{H}$ as long as the values of θ_H and H are not close enough to the point ($\theta_H = \pi/2$; $H = H_u$). When approaching this point, a dramatic change in the direction of $\mathbf{M_0}$ occurs toward the $\mathbf{H}$ direction, and, at a further increase of H, at $\theta_H = \pi/2$ and $H > H_u$, the vector $\mathbf{M_0}$ aligns parallel to the external field.

In our experimental setup (Fig. 1), the susceptibility component χ_h is measured, which is transversal to $\mathbf{H}$. This susceptibility can be considered as a linear response of $\mathbf{M}$ to the applied transversal field dH_h which can be viewed as a response to small change of the direction of the external field H through the angle $dH_h/H = h/H$. In such a case, one gets for the experimental geometry shown in Fig. 1:

$$\chi_h^0 = dM_{0h}/dH_h = (M_0/H)\cos(\theta_H - \theta_0)d\theta_0/\mathrm{d}\theta_H \cong (M_0/H)d\theta_0/d\theta_H, \qquad (5)$$

where the last (approximate) equality is valid close to $\theta_H = \pi/2$. The plots of $\chi_h^0(H/H_u)$ obtained by numerical calculation of Eq. 5 with account made for Eq. 3 at three different θ_H are presented in Fig. 5. It is seen that the position and shape of the calculated curves agree well with the observed pseudoresonance features, with the peak amplitude and quality factor increasing dramatically when approaching the right angle. Note that the derivative in Eq. 5 tends to infinity at $\theta_H \to \pi/2$; in practice, however, this divergence is eliminated due to some inhomogeneous distribution of the $\mathbf{n_u}$ directions. Thus, since the dynamic susceptibility $\chi(\omega)$ should be proportional to χ^0, one can conclude that the observed narrow peak in the r.f. absorption is caused by the abrupt turn of $\mathbf{M_0}$ toward $\mathbf{H}$ when approaching the specific point ($\theta_H = \pi/2$; $H = H_u$).

The next stage is evaluation of the frequency dependence $\chi_h(\omega)$ under the pseudoresonance conditions. To do this, we use again the LLG Eq. 1 in the mode of free evolution ($h = 0$), but now with $\alpha \neq 0$ [13]. Limiting oneself by linear approximation and setting the determinant of the rate equations equal to zero, one

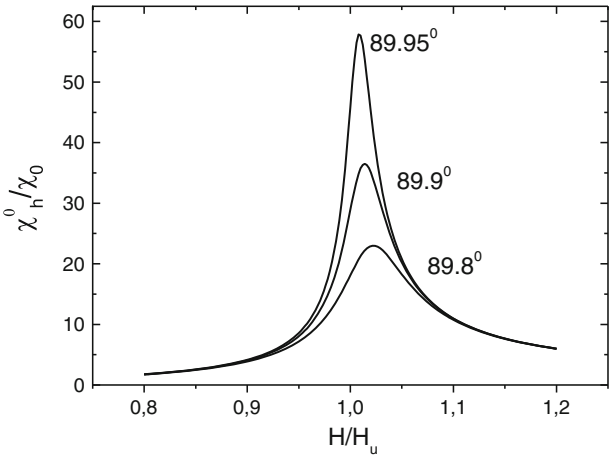

Fig. 5 Field dependences of the transverse static susceptibility (normalized to χ_0) as calculated from Eq. 5 for different angles θ_H indicated at the *curves*

gets the complex eigenfrequency $\omega_c = \omega_c' + i\omega_c''$, where, at the specific point ($\theta_H = \pi/2$; $H = H_u$),

$$\begin{aligned} \omega_c' &= 0; \\ \omega_c'' &= [\alpha/(1+\alpha^2)]\,\gamma(H + 4\pi M_0) \cong \alpha\gamma(H_u + 4\pi M_0). \end{aligned} \tag{6}$$

This solution corresponds to simple exponential relaxation of the magnetization with the characteristic time $\tau = (\omega_c'')^{-1}$, without any oscillations. According to the fluctuation–dissipation theorem, the frequency dependence of the absorption is related to the Fourier transform of the relaxation kinetics and thus reduces to the function similar to the Debye law,

$$\chi''(\omega) = \chi^0 \omega\tau/(1+\omega^2\tau^2) \tag{7}$$

This dependence is shown by the solid curve in Fig. 3, demonstrating a good agreement with the experimental data. The best fit is attained at $\chi^0 = (64 \pm 5)$ (M_0/H) and $\tau = (2.2 \pm 0.3) \times 10^{-9}$ s. As mentioned above, the peak value of χ^0 is limited by inhomogeneity of the film and so can be considered as a free parameter. As to the relaxation time, it should be compared with the prediction of Eq. 6. Using the H_u and M_0 values for the sample under study (see Sect. 2), the fitted value of τ corresponds to $\alpha = 0.006 \pm 0.002$. It is instructive to compare this quantity with the value of $\alpha_{FMR} = 0.003 \pm 0.001$, as extracted from the FMR spectrum measured at 9.5 GHz. Taking into account that the damping parameter may depend on H, the obtained order-of-magnitude agreement seems to be reasonable.

Strictly speaking, the foregoing calculation is justified only for the specific point corresponding to the peak value of r.f. absorption. On either side of this point, the analysis becomes much more complex because of arising of two FMR modes with different non-zero frequencies, see Fig. 4. This problem, related to the shape of the pseudoresonance signal and its behavior in the high-frequency limit, calls for further investigation.

Note that a similar pseudoresonance is expected to arise in other geometry, namely, at the magnetic field **H** directed perpendicular to the film plane (the so-called "easy plane" situation). In this case the specific in-plane uniaxial anisotropy is not required, so the effect might manifest itself in a wide variety of ferromagnetic films, including polycrystalline and even grained ones. This research is planned for the immediate future.

In conclusion, the giant peak of r.f. absorption (magnetic pseudoresonance), which arises in the LSMO thin films with uniaxial plane anisotropy, is found and studied in a wide frequency range of 10–300 MHz. The effect is caused by a steep increase of the transverse magnetic susceptibility which, in its turn, is due to a dramatic change in the orientation of the equilibrium magnetization toward the **H** direction when approaching the specific conditions ($\theta_H = \pi/2$; $H = H_u$). In this point, the FMR frequency equals zero, and the frequency dependence of the pseudoresonance amplitude is found to be of the pure relaxation type, being well described by the expression analogous to the Debye law. Sizeable magnitude and relative ease of detection make this effect favorable for applications in studying magnetic anisotropy and relaxation in nanoscale objects.

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