Ferromagnetic pseudoresonance in anisotropic thin films and its relation to level anticrossing: quantum versus classical

V A Atsarkin, V V Demidov and V Yu Nagorkin
Kotel’nikov Institute of Radio Engineering and Electronics of RAS, 125009 Moscow, Russia
E-mail: atsarkin@cplire.ru

Abstract

Giant peaks of radio-frequency absorption (ferromagnetic pseudoresonance) have been studied at 33 and 290 MHz in the La0.7Sr0.3MnO3 (LSMO) epitaxial thin film with in-plane uniaxial magnetic anisotropy. Unlike previous studies, the pseudoresonance has been registered for both in-plane and out-of-plane orientations of the external magnetic field H0. The observed peaks are attributed to zero-frequency singularities in the spectrum of ferromagnetic resonance (FMR). The data are consistent with classical description based on the free energy and Landau-Lifshits equations. On the other hand, an alternative (quantum) approach is suggested that employs an analogy between the zero-frequency FMR and level anticrossing arising in anisotropic energy spectra of paramagnetic centers when H0 is perpendicular to the symmetry axis. Using the spin-Hamiltonian formalism, we followed the transition from mutual approach of energy levels for spin S → 0 to the level anticrossing at S ~ 10^2 and, finally, to the zero-frequency singularity (ferromagnetic pseudoresonance) at S → ∞. Thus, the transfer is demonstrated from the one-particle (quantum) to many-particle (classical) description.

1. Introduction

Recently, a giant peak of the radio-frequency (r.f.) absorption (the imaginary part of dynamic magnetic susceptibility χ″) was observed in ferromagnetic films which exhibit the in-plane uniaxial magnetic anisotropy [1-4]. It was found that the peak arises when the external magnetic field H0 is in the film plane and directed perpendicularly to the easy anisotropy axis n⊥, whereas its magnitude is equal to the anisotropy field H0 = 2Ku/M0 (here Ku is the first-order term in the uniaxial anisotropy energy and M0 is the saturated magnetization). When the magnitude or direction of H0 passes across this specific point, a narrow peak of χ″ is registered with the width of about 10 Oe and 1 deg., respectively. This feature, however, cannot be considered as conventional ferromagnetic resonance (FMR), since both its position (H0) and shape are independent on the measuring frequency f = ω/2π in a wide r.f. range up to ~10^3–10^9 Hz. As follows from the theory based on the Landau-Lifshits equation [5], the FMR frequency f0 becomes zero in the specific point [1-4], so the FMR is changed to the low-frequency relaxation absorption. For this reason, we will use the term ‘ferromagnetic pseudoresonance’ introduced in [5].

The first study of this phenomenon was performed at the Co-Ni-P films [1]. The peak was attributed to strong increase of static susceptibility due to a steep turn of magnetization when the magnetic field crosses the H0 value. Further, this interpretation was supplemented by consideration of magnetization bistability in the vicinity of the specific point [2]. The peak of r.f. absorption was also observed in a series of epitaxial thin films of La0.7Sr0.3MnO3 (LSMO) prospective for application in nano-electronics [3]; in this case, the effect was employed for accurate determination of the magnitude and direction of the anisotropy field. In the same material, the Debye-like frequency dependence of the peak magnitude was observed, with the relaxation rate of the order of 10^9 s^-1 [4].

In all these studies, only the in-plane arrangement of the magnetic field H0 was used and analyzed. In what follows, we will generalize the consideration and show that the pseudoresonance peaks can be predicted...
theoretically and observed experimentally in the out-of-plane geometry as well, if the field is kept in the plane containing the hard anisotropy axis and the normal to the film. This is reported in section 2 of our communication.

In section 3, an analogy is developed between the zero-frequency ferromagnetic pseudoresonance and the energy level anticrossing which takes place under specific conditions in anisotropic spectra of electron paramagnetic resonance (EPR). Using the spin Hamiltonian formalism with increasing spin S, we will demonstrate the transfer from the quantum description of EPR to classical approach used in ferromagnetism. Thus, one more example will be presented of applicability of the giant spin model developed previously for the treatment of magnetic resonance in superparamagnetic nanoparticles [6–10].

2. Out-of-plane ferromagnetic pseudoresonance

2.1. Theoretical background

Consider a thin ferromagnetic film (in the x-y plane) with the in-plane uniaxial magnetic anisotropy. Let the easy axis \( n_{x} \) is along y, the hard axis is along x, and the anisotropy field is \( H_{a} \). In the presence of external magnetic field \( H_{0} \) with the angular coordinates \((\theta, \varphi)\) (see the inset in figure 1), the FMR frequency \( \omega_{0} \) can be calculated either from the Landau-Lifshits equation or from the expression for the free energy \( U \) [5]. We use the second method. In the case of a thin ferromagnetic film with the in-plane uniaxial anisotropy one has

\[
\frac{U}{M_{0}} = -H_{0}[\sin \theta_{0} \sin \theta \cos (\varphi_{0} - \varphi) + \cos \theta_{0} \cos \theta] + \frac{H_{a}}{2} \left[ 1 - (\sin \theta_{0})^{2} (\sin \varphi_{0})^{2} \right] + 2\pi M_{0} (\cos \theta_{0})^{2}
\]

(1)

where \( \theta_{0}, \varphi_{0} \) are the angular coordinates (polar and azimuthal angles) of the equilibrium magnetization vector \( M_{0} \). The equilibrium orientation of \( M_{0} \) can be found by minimization of equation (1), whereas the FMR frequency reads [5]:

\[
\omega_{0} = \frac{\gamma}{M_{0} \sin \theta_{0}} \sqrt{ \frac{\partial^{2} U}{\partial \theta^{2}} \frac{\partial^{2} U}{\partial \varphi^{2}} - \left( \frac{\partial^{2} U}{\partial \theta \partial \varphi} \right)^{2}} \]

(2)

Numerical calculations performed with the use of equations (1) and (2) showed that the equality \( \omega_{0} = 0 \) which provides a necessary condition for pseudoresonance can be fulfilled only at \( \varphi = 0 \), that is when the field \( H_{0} \) lies in the xz plane. Several \( \omega_{0}(H_{0}) \) diagrams calculated at \( \varphi = 0 \) and various \( \theta \) values with the experimental parameters \( M_{0} = 284 \text{ Oe} \) and \( H_{a} = 206 \text{ Oe} \) (see below) are plotted in figure 1 (the solid curves).

It is seen that the singularities \( \omega_{0} = 0 \) are predicted at specific values \( H_{p} \) of magnetic field which increase from \( H_{a} \) at \( \theta = \pi/2 \) up to \( H_{a} + 4\pi M_{0} \) at \( \theta = 0 \). (The plot at \( \theta = \pi/2 \) was used in previous studies performed with the in-plane geometry [1–4]). These singularities vanish even at minor deviation from the xz plane, see the dashed line plotted at \( \varphi = 2 \text{ deg} \). All diagrams shown in figure 1 consist of two branches (modes) which coincide at \( \omega_{0} = 0 \). The slopes of the low-field and high-field branches near the specific point are very steep and have

\[Figure 1. Calculated FMR frequencies of the ferromagnetic film \( (M_{0} = 284 \text{ Oe}) \) with the in-plane uniaxial anisotropy \( (H_{a} = 206 \text{ Oe}) \) as a function of magnetic field \( H_{0} \) directed at the polar angles \( \theta \) (indicated at the curves) and azimuthal angles \( \varphi = 0 \) (solid lines) and \( \varphi = 2 \text{ deg} \) (dashed line). The high-field parts of the diagrams are not shown. Inset: sketch of the experimental geometry.\]
Our experiments were performed with the epitaxial thin film of ferromagnetic manganite La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO) with the Curie temperature $T_C$ of about 350 K. This material was thoroughly explored in the last decades [11]. The LSMO film of 50 nm thick was grown by laser ablation on the (110) surface of the NdGaO$_3$ (NGO) substrate. It is known [12] that the basic plane of the LSMO films grown on the orthorhombic (110) NGO substrate is (011) (we use the pseudocubic notation for LSMO), and an additional in-plane uniaxial magnetic anisotropy is created with the easy axis directed along [010] LSMO due to orthorhombic distortions of the LSMO crystal structure. In the samples under study, the substrate plates of the $5 \times 5 \times 0.5$ mm$^3$ were cut in such a way that the LSMO easy axis (corresponding to the [110] direction of the NGO substrate) coincided with an edge of the substrate.

The sample used in our experiments was chosen from the series of LSMO/NGO epitaxial films described in [3, 4]. The pseudoresonance feature was observed in all samples of this series. For the main set of our measurements, the best-quality film was chosen. The selection criterion was the minimal width ($\Delta H$) of the FMR line registered by standard techniques (see below). The experiments described in the next subsection were performed with the film revealing the room-temperature FMR line-width of about 20 Oe for the in-plane arrangement of magnetic field. This value could be compared with the minimal line-width $\Delta H = 12$ Oe attained up to day at similar LSMO/NGO films and caused mostly by the natural (homogeneous) damping [13]. The extra broadening characterizes the inhomogeneity of our sample.

The main magnetic parameters of the sample such as $M_{ho}$ $H_0$ and the direction of the anisotropy axis $n_0$ were determined from the angular dependence of the standard FMR spectrum registered at the in-plane arrangement of $H_0$ at room temperature [3]. The commercial X-range EPR spectrometer Bruker ER 200 was used. Along with the uniaxial anisotropy, the natural crystallographic cubic anisotropy was revealed. At room temperature, however, the anisotropy field $H_0$ of the sample under study amounted to 206 Oe and exceeded strongly the natural cubic-symmetry anisotropy field of about 10 Oe, so the latter was disregarded.

The measurements of the r.f. absorption were performed at room temperature. Two home-made r.f. spectrometers based on the Q-meter principle and working at the frequencies $\omega/2\pi = f = 290$ MHz and 33 MHz were used. According to the previous (in-plane) results [4], this frequency range overlaps the central part of the Debye-like frequency dependence of the absorption peak magnitude. The sample was placed in the central area of the r.f. pickup coil and can be rotated about two orthogonal axes with the accuracy of $\pm 0.1$ deg. with the use of the high-precision goniometer. The mutual arrangement of the static and r.f. magnetic fields, anisotropy direction and coordinate axes is shown in figure 1 (inset). In fact, the $H_0$ direction was fixed in horizontal position, whereas the angles $\theta$ and $\varphi$ were controlled by the rotation of the sample. In the main set of measurements, the sample was rotated about the vertical axis coinciding with $n_0$, so $H_0$ was kept in the $xz$ plane ($\varphi = 0$). The signals were recorded upon sweeping the magnetic field through the peak position. To increase the sensitivity, accumulation was used. The measuring r.f. field $h$ was directed vertically. Proportionality between the absorption and incident r.f. power was observed within the $h$ range up to 2 mOe.

### 2.3. Experimental results and discussion

Experimental data are presented in figure 2. Typical absorption peaks are shown in figure 2(a); figure 2(b) shows the angular dependence of the pseudoresonance field $H_p(\theta)$ at $\varphi = 0$. One can state that the pseudoresonance peaks really exist at every direction of $H_0$ in the $xz$ plane. It is seen that the peak position $H_p$ does not depend on frequency within the range of 33–290 MHz and agrees very well with the theoretical curve calculated by the use of equations (1), (2). It should be noted that the pseudoresonance peak broadens as the field $H_0$ is deflected from the film plane. This is obviously related to the progressive raising of the angle between two FMR branches around $H_p$ when $\theta$ decreases from $\pi/2$ to 0, see figure 1. At the extreme position, when $H_0$ is perpendicular to the film, the mutual deviation of the branches becomes sufficient to provide partial resolution of the peak at $f = 290$ MHz into two components, thus demonstrating the transition from pseudoresonance to conventional FMR. Unlike that, the peak remains unresolved at 33 MHz. Note that the maximum absolute value of the absorption coefficient ($\chi''$) was estimated as about 15$M_0/H_0$ for $f = 290$ MHz and $\theta = \pi/2$. Finally, the peak broadens and disappears when the field $H_0$ is deflected from the $xz$ plane through an angle $\varphi$ exceeding $\sim 1$ deg., see figure 2(c). Thus, the theoretical predictions (section 2.1) have been totally confirmed.
3. Level anticrossing and ferromagnetic pseudoresonance: quantum versus classical

In this part of our communication we leave aside the details of the experimental data and concentrate on physical origin of the ferromagnetic pseudoresonance in more general aspect. The main point of our consideration is the analogy between zero-frequency points \( \omega_0 = 0 \) in the FMR spectrum and crossing (really, anticrossing) of energy levels that occurs under some specific conditions in spectra of paramagnetic centers. In the latter case, the resonance frequency is determined by the energy difference of the converging levels and so can fall into the low-frequency range not typical for conventional electron paramagnetic resonance (EPR). In what follows, we will demonstrate the transfer between these phenomena and reveal correspondence between quantum and classical approach in description of spin systems.

Consider non-interacting paramagnetic centers with spin \( S \) subjected to an external magnetic field \( H_0 \) and crystalline field with uniaxial symmetry. The energy spectrum of such a system is determined by the spin Hamiltonian (in frequency units) [14, 15]:

\[
\frac{\mathcal{H}}{\hbar} = \gamma (H_0 \cdot \mathbf{S}) + D \left[ S_Z^2 - \frac{S(S+1)}{3} \right]
\]

where \( \gamma < 0 \) is the magnetogyric ratio, \( D \) is the parameter related to the axial anisotropy (hereafter we suppose \( D < 0 \)), and \( Z \) is directed along the anisotropy axis. Let \( H_0 \) be perpendicular to the crystal axis and, to start with, \( S = 3/2 \) (for \( \text{Cr}^{3+} \) ion, for example). The well-known energy diagram is shown in figure 3(a), where the magnetic field \( H_0 \) and energy \( E \) are normalized to the anisotropy field \( H_a \) and initial (zero-field) splitting \( \Delta_0 = |D(2S - 1)| \), respectively. Here we make use of the relation

\[
\gamma H_a = \Delta_0
\]
suggested in the framework of the giant spin model [6–8]. In this model, the FMR spectrum is associated with the lowest states of the high-spin multiplet and equation (4) is justified by comparison of the quantum (paramagnetic) and classical (ferromagnetic) expressions for the anisotropy energy.

As seen in figure 3(a), the levels 1 and 2 practically coincide at low enough external fields. When the field increases, the gap $\omega_{12}$ between the levels 2 and 3 decreases and passes through a minimum denoted as $\Delta_{LAC}$ (marked by an arrow) at $H_0 = H_{LAC}$. This is just the level anticrossing (LAC) to be considered. One may argue that the levels 2 and 3 are still far from real crossing, so the energy difference is rather large. However, the crossing approaches upon increasing of the spin value $S$, see below. Both the (2, 3) and (1, 2) level pairs diverge at further increasing of the external magnetic field. The inversion of the level slopes implies a strong change in quantum states and a steep turn of magnetic moment. As a result, the corresponding component of magnetic susceptibility increases considerably under the LAC conditions.

Let us follow the evolution of the LAC feature upon increasing $S$ up to the values typical of superparamagnetic behavior. The further step to ferromagnetism implies thermal depopulation of all energy levels of the spin multiplet except the lowest ones. Thus, we will be interested in the three lowest energy levels only. The dependences of the frequencies $\omega_{12}$ and $\omega_{23}$ on the external magnetic field $H_0 = H_{LAC}$ have been calculated with the spin Hamiltonian equation (3) at $S = 10, 20, 40, 80$ and $200$. The results are presented in figure 3(b). When $S$ increases, the LAC peculiarity becomes more pronounced: the value of $\Delta_{LAC}$ decreases progressively and apparently tends to zero, whereas $H_{LAC}$ approaches the value of $H_n$. Note that the degeneracy of the (1, 2) doublet is removed at the same point.

Passing to the ferromagnetic limit ($S \to \infty$), one can expect that the FMR spectrum should be formed by the transition from the only populated lowest level to the next one, that is (1, 2) $\to$ 3 at $H_0 < H_{LAC}$ and 1 $\to$ 2 at $H_0 > H_{LAC}$. At $H_0 = H_{LAC} = H_n$ the FMR frequency tends to zero. This singularity along with a steep change in the quantum states of the involved levels at the LAC point should provide a strong increase of both static and dynamic magnetic susceptibility (magnetic pseudoresonance).

The same result can be obtained if one starts from the opposite side using the classical FMR approach. As an analogue of the above system one can consider a ferromagnetic sphere with the uniaxial magnetic anisotropy $K_u$ in the external magnetic field $H_0$ directed perpendicularly to the easy axis $n_0$. The dependence of the FMR frequency $\omega_{0}$ on $H_0$ can be calculated using Landau–Lifshits equation or differentiation of the free energy as it was done in section 2.1 [5]. Similar result is obtained for ferromagnetic film with the in-plane anisotropy and $H_0$ directed along the hard axis ($\theta = \pi/2; \varphi = 0$), but in this case the zero-field splitting changes to $\gamma^2\mu_B(H_0^2 - 4\pi M_0)$ due to the demagnetizing field. The calculated curve is shown in figure 3(b) with the thick solid line. Evidently, the $\omega_{0}(H_0)$ curve calculated with the standard FMR methods looks as the limit of the paramagnetic analogues at $S \to \infty$; this confirms the validity of our approach. It is of interest that the low-field

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**Figure 3.** (a) Energy diagram for paramagnetic centers with $S = 3/2$ in the external magnetic field $H_0$ directed perpendicularly to the crystal symmetry axis. The level anticrossing is indicated by an arrow. (b) The transition frequencies $\omega_{12}$ (thin solid lines) and $\omega_{23}$ (thin dashed lines) for paramagnetic centers with $S = 10, 20, 40, 80$ and $200$ (from left to right) at $H_0$ directed perpendicularly to the crystal symmetry axis. The thick solid line shows the FMR frequency of ferromagnetic film with uniaxial anisotropy at $H_0$ directed along the in-plane hard axis.
and high-field branches of the $\omega_0(H_0)$ diagram correspond to different quantum transitions $\omega_{23}$ and $\omega_{12}$, respectively.

4. Conclusion

In conclusion, the phenomenon of great increase of low-frequency susceptibility in anisotropic ferromagnetic films (ferromagnetic pseudoresonance) is studied within more general approach including both the in-plane and out-of-plane geometry. It is shown that the pseudoresonance peaks associated with zero FMR frequency can be observed if the external magnetic field $H_0$ lies in the plane containing the hard anisotropy axis and the normal to the film surface. The theoretical predictions are confirmed in the experiments performed at the epitaxial LSMO film grown on the NGO substrate. Excellent agreement is achieved between the experimental and predicted angular dependencies of the peak field $H_p$.

Reasoning from an analogy between the zero points in the FMR frequency diagram and energy level anticrossing occurring in the EPR spectra, the transfer is demonstrated from the low-spin quantum description based on the spin Hamiltonian to classical approach used in FMR at $S \rightarrow \infty$. The transition between quantum and classical descriptions is of fundamental physical interest, being intimately coupled with conversion from the one-particle to many-particle problem.

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ORCID iDs

V A Atsarkin  https://orcid.org/0000-0002-3937-9412

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