ISSN 2072-5981

Magnetic Resonance in Solids

Electronic Journal

Volume ***20,***
Issue ***1***
Paper No ***18104,***
1-8 pages
2018

http://mrsej.kpfu.ru
http://mrsej.ksu.ru

Established and published by Kazan University
Sponsored by International Society of Magnetic Resonance (ISMAR)
Registered by Russian Federation Committee on Press, August 2, 1996
First Issue was appeared at July 25, 1997

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"***Magnetic Resonance in Solids. Electronic Journal***" (**MRSej**) is a peer-reviewed, all electronic journal, publishing articles which meet the highest standards of scientific quality in the field of basic research of a magnetic resonance in solids and related phenomena.

Indexed and abstracted by
Web of Science (*ESCI*, Clarivate Analitics, from 2017), *Scopus* (Elsevier, from 2012), *RusIndexSC* (*eLibrary*, from 2006), *Google Scholar*, *DOAJ*, *ROAD*, *CyberLeninka* (from 2006), *SCImago Journal & Country Rank*, etc.

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

Degenerated nuclear magnetostatic modes in ferromagnets

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(Received April 6, 2018; revised May 8, 2018; accepted May 10, 2018; published May 23, 2018)

The detailed calculations of dependence of the frequency of oscillations on the external magnetic field and on the shape of ferromagnet sample with interacting electronic and nuclear subsystems were done. Such system possesses a discrete number of eigenfrequencies of precession of magnetization. It is shown that these magnetostatic eigenmodes can be degenerated, i. e. different modes can have the same frequency. The dependence of these degenerated modes on the sample shape is investigated. It is shown that the properties of magnetization precession and even the number of eigenmodes depend on the external magnetic field and on the shape of the sample. These effects may produce specific features on the NMR spectra of magnetic materials.

PACS: 76.60.-k, 75.78.-n

Keywords: Electron-nuclear interaction, magnetostatic boundary conditions, NMR

1. Introduction

Nuclear magnetic resonance (NMR) is one of the most effective methods of investigation of local electronic and spin density distribution [1]. But analysis of NMR signals from magnetic ordered systems is a quite complicated problem because of some specific features. Firstly, NMR frequency in magnetic solids is defined mainly by local magnetization, the value of which can differ significantly from external magnetic field. Secondly, resonance transitions of nuclear spins are induced mainly by variable part of electronic magnetization. Small perturbating field produces quite a big oscillations of hyperfine field, which is proportional to the dynamic part of electronic susceptibility at the NMR frequency. It is the effect of amplification of magnetic field (the value of amplification factor usually is about $10 - 10^3$). Thirdly, at the low temperatures the indirect Suhl-Nakomura interaction (SNI) between nuclear spins plays an important role in magnetic dynamics [2]. It effects in substantial broadening of NMR line and in dynamic frequency shift (pulling). The value of pulling depends on the temperature and on the amplitude of nuclear magnetization. The broadening of NMR line and frequency pulling produce specific features in the system dynamics. For example, as it was shown in [3,4], the relaxation in nuclear subsystem can be produced through relaxation in electronic subsystem. In this case the absolute value of nuclear magnetization remains constant and hence, the relaxation differs from the Bloch type. In addition, the dependence of NMR frequency in magnets on the amplitude of nuclear magnetization determines nonlinear properties of the nuclear spin system. It leads to appearance of specific characteristics in NMR absorption and dispersion signals [4]. SNI also can effect in correlated motion of nuclear spins (nuclear spin oscillations and waves).

In this paper we are interesting in two problems. The first problem is the interaction between electronic and nuclear subsystems in ferromagnet sample and the second one is the effect of boundaries of the sample (and demagnetization factors, in particular) to the magnetic properties of the system. The investigation of interaction between different subsystems in ferromagnets and ferromagnet based layered structures is one of the modern direction in the field of magnetic phenomena physics. For example, the book [5] is devoted to the theoretical investigation of interaction between acoustic and spin waves. Here the interaction between electronic and nuclear system is studied. In this case amplification factor, frequency pulling, relaxation and other NMR characteristics are defined by the spectrum and by the dynamic susceptibility of electronic subsystem. If we deal with samples of finite size, the boundary effects and demagnetization factors change significantly the dynamic properties of electronic subsystem. The Maxwell equations have to be taken into account to describe this situation. But in the case of slow variable magnetic oscillation at the NMR frequency this equations can be used in magnetostatic approximation with corresponding boundary conditions. The boundary effects produce specific properties of the magnetostatic waves propagating in the system and a number of papers are devoted only to the investigaion of the properties of electronic subsystem. For example, some manuscripts are devoted only to the investigation of spectrum of magnetostatic wave in ferromagnet films and layered structures [6]. Some other works are devoted entirely to the investigation of the properties of ferromagnetic resonance (FMR) in different magnetic media (ferro- and antiferromagnets, ferrites, layered structures, etc.) [7]. Even the study of the FMR linewidth in the sample of finite size (in particular, in layered structures) is a quite complicated problem [8]. The another effect of boundaries is the appearance of discrete number of additional magnetostatic modes near the ferromagnetic resonance (FMR) frequency. It is brightly expressed in the case of samples, limited in all three dimentions, in particular, in spherical samples [9]. The spectrum of electronic subsystem has additional maxima. This should be manifested in appearance of fine structure in frequency pulling in nuclear subsystem. In other words, the nuclear magnetostatic modes can be observed. The first experemental observation of such modes was done in 1966 [10] and first theoretical description was suggested in 1967 [11] for the nuclear magnetostatic modes of Mn^{55} in $MnFe_2O_3$ compound with spinel structure. Further such effects have been observed also in two-sublattice antiferromagnet MnF_2 [12].

In this paper we present the results of theoretical investigation of coupled precession of electronic and nuclear magnetization in the samples of spheroidal shape. We show that eigenmodes of precession of magnetization have a fine structure. For the samples with some shape in the external magnetic field these eigenmodes can be divided into two well separated groups, first of them can be considered as nuclear eigenmodes, and second as electronic ones. For other samples and magnetic fields nuclear and electronic eigenmodes become mixed. In addition, it was shown [3, 4] that at low temperatures some NMR effects, produced by homogeneous oscillations of nuclear magnetization can be observed. This paper shows, that the precession of nuclear magnetization may also demonstrate complicated structure and may have interesting specifics if we deal with the samples of the finite size.

2. Model

Lets consider ferromagnet sample of spheroidal shape. Suppose also that all nuclei have magnetic moment. The energy of the system can be written as

$$\begin{aligned} F = \hat{A}\mathbf{M}\mathbf{m} - M_z H_A - H_0(M_z + m_z) - h_x(M_x + m_x) - h_y(M_y + m_y) \\ + \frac{1}{2}[N_x(M_x + m_x)^2 + N_y(M_y + m_y)^2 + N_z(M_z + m_z)^2]. \end{aligned} \tag{1}$$

Here $\mathbf{M}$ and $\mathbf{m}$ are the electronic and nuclear magnetization. The first term in Eq. (1) describes the hyperfine interaction between electronic and nuclear spins, the second one is the easy-axis anisotropy energy. The interaction with external constant $\mathbf{H}_0 = (0, 0, H_0)$ and variable

$\mathbf{h} = (h_x, h_y, 0)$ magnetic field is taken into account. The last term is the demagnetization energy of ellipsoid. The sample under investigation is supposed to be of spheroidal shape. The boundary of the sample is defined as

$$\frac{x^2+y^2}{a^2} + \frac{z^2}{b^2} = 1. \tag{2}$$

It is convinient to use shape parameter $\alpha = a/b$. We deal with oblate spheroid if $\alpha > 1$, and with oblong one if $\alpha < 1$. The expressions for demagnetization factors $N_{x,y,z}$ for spheroids depend on the parameter α and can be found in [13]. It follows from the symmetry that $N_x = N_y = (4\pi - N_z)/2$ for spheroids ($\alpha \neq 1$) and $N_x = N_y = N_z = 4\pi/3$ for sphere ($\alpha \to 1$).

The motion of magnetization is described in terms of Maxwell equations, but in the case of frequencies at the vicinity of NMR the magnetostatic approximations can be used:

$$\begin{aligned} &[\nabla, \mathbf{h}] = 0, \\ &(\nabla, \mathbf{h} + 4\pi(\mathbf{M}+\mathbf{m})) = (\nabla, \mathbf{h} + 4\pi\hat{\chi}\mathbf{h}) = (\nabla, \hat{\mu}\mathbf{h}) = 0, \end{aligned} \tag{3}$$

where $\hat{\chi}$ is the magnetic susceptibility tensor and $\hat{\mu}$ is the permeability tensor. The solution of first equation (3) is $\mathbf{h} = \nabla\psi$, where ψ is the magnetostatic potential, and the second one gives Walker equation [9]

$$\mu\left(\frac{\partial^2\psi}{\partial x^2} + \frac{\partial^2\psi}{\partial y^2}\right) + \frac{\partial^2\psi}{\partial z^2} = 0. \tag{4}$$

Here $\mu = 1 + 4\pi\chi_{xx}$ and $i\mu_1 = \chi_{xy}$ are diagonal and off-diagonal components of permeability tensor, which can be obtained by solving the system of linearized Landau-Lifshitz and Bloch equations for electronic and nuclear magnetization

$$\frac{d\mathbf{M}}{dt} = -\gamma_e[\mathbf{M}, \mathbf{H}_\mathbf{M}], \qquad \frac{d\mathbf{m}}{dt} = \gamma_n[\mathbf{m}, \mathbf{H}_\mathbf{m}], \tag{5}$$

where γ_e and γ_n are electronic and nuclear gyromagnetic ratios (the signs of gyromagnetic ratios are taken into account in Eq. (5), hence $\gamma_e > 0$, $\gamma_n > 0$ in our notations), and

$$\mathbf{H}_\mathbf{M} = -\frac{\partial F}{\partial \mathbf{M}}, \qquad \mathbf{H}_\mathbf{m} = -\frac{\partial F}{\partial \mathbf{m}} \tag{6}$$

are effective magnetic fields acting on the electronic and nuclear magnetization respectively. We neglect the Hilbert and Bloch dissipation terms in Eq. (5) for simplicity. The expressions for μ and μ_1 in the linear approximation contain two additive contributions from electronic and nuclear magnetization subsystems and they are not shown here because of their unwieldy. It is important that effective fields $\mathbf{H}_\mathbf{M}$ and $\mathbf{H}_\mathbf{m}$ depend on the demagnetization factors $N_{x,y,z}$. That is why the components of permeability tensor μ and μ_1 depend on the $N_{x,y,z}$, which, in turn, depend on the shape parameter α. In other words, we take into account the shape of the sample through the demagnetization factors.

Outside the sample we have $\mu = 1$, $\mu_1 = 0$ and Walker equation (4) transforms to the Laplace one. Next we will construct solution of Eq. (4) inside and outside the sample, supposing magnetostatic potential ψ to be limited everywhere and equal to zero at the infinity. The magnetostatic boundary conditions require the continuousness of the tangential components of magnetic field $\mathbf{h} = \nabla\psi$ and normal components of magnetic induction $\mathbf{b} = \mathbf{h} + 4\pi(\mathbf{M}+\mathbf{m})$ at the boundary of the sample. It leads to the dispersion equation, which is constructed as follows.

The solution of Laplace equation outside the sample can be written as

$$\psi^{\text{out}} = e^{im\phi}(ar)^{-n-1}P_n^{|m|}(\cos(\theta)), \quad a/b = 1, \tag{7}$$
$$\psi^{\text{out}} = e^{im\phi}Q_n^m(i\xi)P_n^{|m|}(\eta), \quad a/b > 1, \tag{8}$$
$$\psi^{\text{out}} = e^{im\phi}Q_n^m(\xi)P_n^{|m|}(\eta), \quad a/b < 1, \tag{9}$$

where P_n^m and Q_n^m are the Legendre functions of the first and second kind, r, θ, ϕ are the spherical coordinates, and ξ, η, ϕ are the spheroidal coordinates, which are defined as

$$x = \frac{d}{2}\sqrt{\xi^2+1}\sqrt{1-\eta^2}\cos\phi, \quad y = \frac{d}{2}\sqrt{\xi^2+1}\sqrt{1-\eta^2}\sin\phi, \quad z = \frac{d}{2}\xi\eta, \quad a/b > 1, \tag{10}$$
$$x = \frac{d}{2}\sqrt{\xi^2-1}\sqrt{1-\eta^2}\cos\phi, \quad y = \frac{d}{2}\sqrt{\xi^2-1}\sqrt{1-\eta^2}\sin\phi, \quad z = \frac{d}{2}\xi\eta, \quad a/b < 1 \tag{11}$$

for oblate and oblong spheroid correspondingly, $d = \sqrt{|a^2-b^2|}$ is the distance between two foci. The boundary of the sample is defined as $r = 1$ for the case of spherical sample and as $\xi = \xi_0 = \dfrac{1}{\sqrt{|\alpha^2-1|}}$ for spheroids. The solution of Walker equation inside the sample can be written as follows

$$\psi^{\text{in}} = C_{nm}e^{im\phi}\int\limits_{-\pi}^{\pi} P_n\left(\beta r(\sqrt{-\mu}\cos\theta + \sin\theta\cos u)\right)\cos(mu)\,du, \tag{12}$$

with $\beta = -(1-\mu)^{-1/2}$ for the spherical sample and

$$\psi^{\text{in}} = C_{nm}e^{im\phi}\int\limits_{-\pi}^{\pi} P_n\left(\beta(-\sqrt{-\mu}\xi\eta + \sqrt{1+\xi^2}\sqrt{1-\eta^2}\cos u)\right)\cos(mu)\,du, \tag{13}$$

with $\beta = (1-\xi_0^2(\mu-1))^{-1/2}$ for oblate spheroid. The same formula (13) is used for oblong spheroid, but one should change $\sqrt{1+\xi^2} \to \sqrt{\xi^2-1}$ and $\beta = (-1-\xi_0^2(\mu-1))^{-1/2}$ in this case. The continuousness of tangential components of magnetic field $\mathbf{h} = \nabla\psi$ at the boundary $\xi = \xi_0$ (or $r = 1$) requires the coefficients

$$C_{nm} = \frac{1}{2\pi}\frac{(n+|m|)!}{(n-|m|)!}\frac{K}{P_n^m(S)}, \tag{14}$$

where $K = 1$ for $\alpha = 1$, $K = Q_n^m(i\xi_0)$ for $\alpha > 1$ and $K = Q_n^m(\xi_0)$ for $\alpha < 1$. The continuousness of normal components of magnetic induction $\mathbf{b} = \hat{\mu}\mathbf{h}$ gives the characteristic equation

$$L(\xi_0) + S\frac{d}{dS}\ln(P_n^{|m|}(S)) - \frac{\mu_1 m}{\alpha^2} = 0, \tag{15}$$

where $L(\xi_0) = n + 1$ for the case of sphere ($\alpha = 1$), $L(\xi_0) = -i\xi_0\dfrac{d}{d(i\xi_0)}\ln(Q_n^{|m|}(i\xi_0))$ and $L(\xi_0) = -\xi_0\dfrac{d}{d\xi_0}\ln(Q_n^{|m|}(\xi_0))$ for the case of oblate ($\alpha > 1$) and oblong ($\alpha < 1$) spheroid. The new variable S is defined as $S = S(\omega) = -\dfrac{\sqrt{-\mu}}{\sqrt{\alpha^2-\mu}}$. Note, that S locates in the $(-1, 0)$ interval if $\mu < 0$, in the $(-\infty, -1)$ region if $\mu > \alpha^2$, and S is imaginary if $0 < \mu < \alpha^2$. All these three cases should be considered separately because of the specifics of calculation of Legendre functions $P_n^m(S)$. Note, that the dependence on shape parameter in Eq. (15) is twofold. It is the explicit dependence: S depends on α, and the implicit one, because μ and μ_1 are the functions of α through the demagnetization factors. The solution of characteristic equation (15) gives the eigenfrequencies of coupled oscillations of electronic and nuclear magnetization. Eq. (15) has the three-indexed solution. First two indexes n, m follow from the boundary problem. The finite solution corresponds to the indexes $n = 1, 2, \ldots$ and $m = 0, 1, \ldots n$. Third index r numerates the roots of Eq. (15) with given (n, m). The number of roots depends both on the (n, m) index and the magnetic field H_0 and the shape parameter α. In particular for the simplest eigenmode $(n, m) = (1, 1)$ Eq. (15) for usual experimentally used magnetic field $H_0 \sim 3 \div 10\,\mathrm{kOe}$ and for samples close to sphere has two well separated solutions, one of which refers to the electronic oscillations and another to the nuclear oscillations.

3. Results

The structure of solutions of characteristic equation (15) depends on the value of magnetic field H_0 and on the shape parameter α. For some values (H_0, α) the eigenfrequencies of nuclear and electronic modes are close to each other and it is difficult to separate one mode from another. Such situation is demonstrated at Fig. 1 where the field dependence of lowest magnetostatic modes in spherical sample is shown. For modes $(n, m) = (1, 1), (2, 0), (2, 1), (2, 2)$ characteristic equation (15) has two solutions if magnetic field H_0 exceeds some value $(H_{n,m})_{\mathrm{crit}}$. The lowest branch represents nuclear mode and the highest is refered to the electronic one. Mode with $(n, m) = (6, 0)$ has six branches: three electronic modes and three nuclear modes. The third index r is requaried to numerate the different eigenfrequencies. To construct Fig. 1 and the next pictures the known parameters for ferromagnet $\mathrm{MnFe_2O_4}$ were used: saturation magnetization is equal to $M_0 = 560\,\mathrm{Oe}$, hyperfine fields are $AM_0 = 586\,\mathrm{kOe}$ and $Am_0 = 8\,\mathrm{Oe}$ (hence, hyperfine constant is $A = 1046$ and nuclear magnetization $m_0 = 7.6 \cdot 10^{-3}\,\mathrm{Oe}$), anisotropy field is $H_a = 1\,\mathrm{kOe}$ and gyromagnetic ratios are $\gamma_e = 17.58\,\mathrm{GHz/kOe}$, $\gamma_n = 6.28 \times 10^{-3}\,\mathrm{GHz/kOe}$ [14].

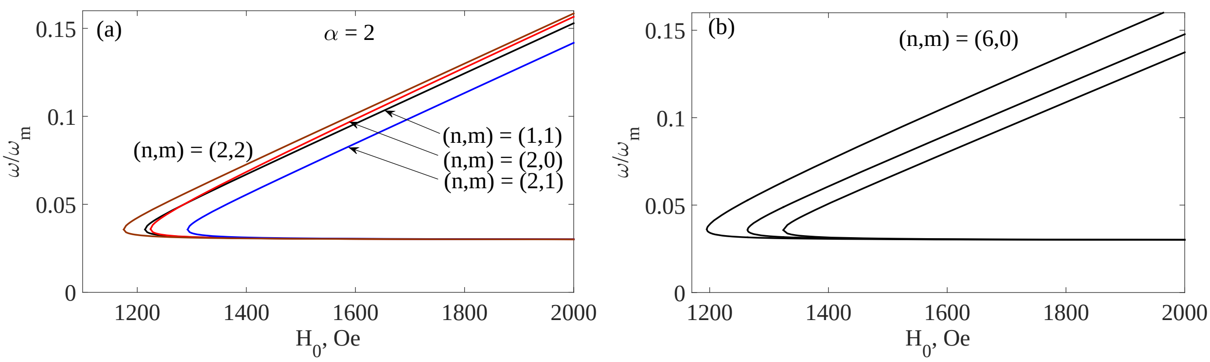

Figure 1. The dependence of electronic and nuclear magnetostatic modes on external magnetic field H_0 for $(n, m) = (1, 1), (2, 0), (2, 1), (2, 2)$ (a) and for $(n, m) = 6, 0$ (b)

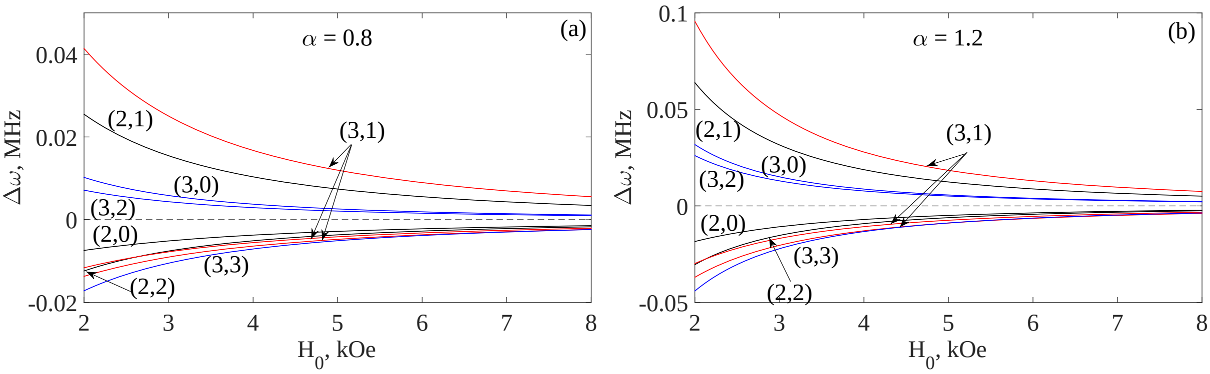

Figure 2. The separation of lowest nuclear oscillation modes from $(1, 1)$ eigenmode versus external magnetic field

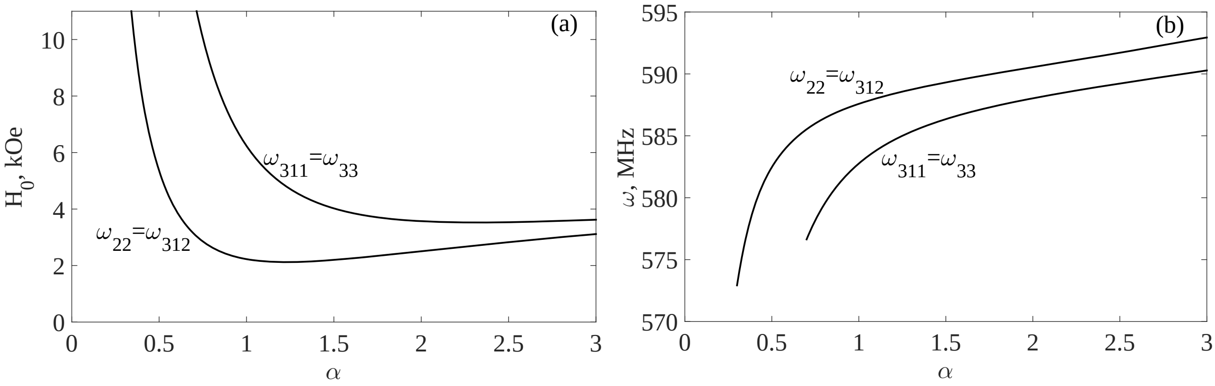

Figure 3. Dependence of magnetic field (a) and degenerated frequencies (b) on the shape parameter of the sample

As can be seen from Fig. 1 the number of possible eigenmodes and their behavior depends strongly on the magnetic field, especially if $H \sim H_{\text{crit}}$. The eigenmode with index (n, m) does not exist if the value of magnetic field H_0 less than H_{crit}. The critical field H_{crit} depends on the indices (n, m, r) and can be calculated numerically. The investigation of specific properties of the spectrum near this critical region is outside the frames of this paper. Now we restrict ourselves in the situation of quite a big field H_0, when nuclear and electronic modes are well separated and we will take into account only nuclear modes. The simplest eigenmode has index $(n, m) = (1, 1)$. It is convenient to compare all other eigenmodes with $(1, 1)$. The separation $\Delta\omega = \omega_{n,m} - \omega_{1,1}$ of lowest nuclear magnetastatic modes as a function of magnetic field is shown in Fig. 2 for $\alpha = 0.8$ (oblong spheroid) and $\alpha = 1.2$ (oblate spheroid).

For $(n, m) = (2, 0)$, $(2, 2)$, $(3, 0)$, $(3, 2)$ and $(3, 3)$ only one nuclear eigenmode exists ($r = 1$), and for $(n, m) = (3, 1)$ we have three eigenmodes ($r = 1, 2, 3$). So, the total number of nuclear eigenmodes with $n \leq 3$ is equal to 9. Some specific features can be seen from Fig. 2. Firstly, the frequency separation between eigenmodes decreases with the magnetic field H_0. Secondly, the separation for the same field for oblong spheroid is smaller than for oblate spheroid. Thirdly, some eigenmodes cross each other. In particular, mode $(n, m) = (2, 2)$ crosses mode $(n, m, r) = (3, 1, 2)$ and mode $(n, m, r) = (3, 1, 1)$ crosses mode $(n, m) = (3, 3)$. Note, that crosspoint of the modes means the degenerating of eigenfrequencies: two different eigenmodes has the same frequency in the same external magnetic field H_0. If in addition we take into account modes with $n = 4$ and $m = 0, 1, 2, 3, 4$ the total number of modes will be equal to 19 and the number of crosspoints at the region of interest ($\alpha \sim 1$, $H_0 \sim 1 \div 10\,\text{kOe}$) increases up to 10. But it is obvious, that the structure of magnetic potential ψ becomes more complicated with the increasing of number n (see (7)-(9) and (12),(13)). So, the structure of internal field $\mathbf{h} = \nabla\psi$ and magnetization $\mathbf{M} = \hat{\chi}\mathbf{h}$ also becomes complicated. It seems that such structures can be hardly observed in experiment and that is why we will not consider here the highest modes. But we investigated in detail the crosspoints of modes $(2, 2)$-$(3, 1, 2)$ and $(3, 1, 1)$-$(3, 3)$. If the sample has spheroidal shape with given parameter $\alpha = a/b$ the crosspoint of modes occurs at some fixed magnetic field H_0. If the sample with another α is used, the crosspoint will be at another value of magnetic field. The dependence of this field of degenerating on the shape parameter α is shown at Fig. 3 (a). We took into account region of magnetic field $H_0 \sim 2 - 10\,\text{kOe}$, which usually used in NMR experiments. The method of using the Fig. 3 (a) is the follows. If we have the sample with given α we can find the magnetic degenerating field H_0. One curve at Fig. 3 (a) corresponds the $\omega_{22} = \omega_{312}$ degeneration, and another one – the $\omega_{311} = \omega_{33}$ degeneration. The correspond eigenfrequency can be found from Fig. 3 (b). Of couse these eigenfrequencies are about $\omega \sim 580\,\mathrm{MHz}$ and they are close to the frequency of Mn^{55} nuclei in $MnFe_2O_4$ compound [1]. Note, that according to our model, the characteristic equation for eigenfrequencies has different form for the case of oblong spheroid ($\alpha < 1$) sphere ($\alpha = 1$) and oblate spheroid ($\alpha > 1$). But the solution of this equation passes trough the point $\alpha = 1$ smoothly and continuously. The result is entirely predictable, because it is obvious, that the case of the spherical sample is not the special from physical point of view.

4. Summary

The coupled oscillations of electronic and nuclear magnetization in spheroidal ferromagnet samples have interesting specific features. The characteristic equation for eigenfrequencies of precession of magnetization which takes into account magnetostatic boundary conditions has three different forms for the cases of oblong spheroid ($\alpha < 1$), sphere ($\alpha = 1$) and oblate spheroid ($\alpha > 1$), but all dynamic characteristics of this precession change continuously with α. Since we deal with three dimensional problem, the eigenvalues are given by three indices: n, m and r, where $n = 1, 2, \ldots$ and $m = 0, 1, \ldots n$ follow from magnetostatic boundary conditions and r is the root number of characteristic equation. The number of possible modes depends on the external magnetic field and on the shape parameter. Each eigenmode has it's own dependence on the magnetic field. Different eigenmodes can cross each other at the special value of magnetic field. Two different eigenmodes have the same eigenfrequency at this field of degenerating. The dependence of field of degenerating of lowest eigenmodes on the shape of sample have been studied. It was done using numerical analysis of the solutions of characteristic equation. The corresponding frequencies of degenerated eigenmodes are also the functions of shape parameter. They also have been obtained numerically. We investigated the dependence of eigenmodes on the sample shape in this paper, but the alternative interpretation of our results is also possible. The values μ, μ_1 in characteristic equation depend on demagnetization factors $N_{x,y,z}$, where $N_x = N_y = (4\pi - N_z)/2$ according to the symmetry. But the demagnetization factors are the monotonic functions of the shape parameter α, namely N_z changes from 0 to 4π when α changes from 0 to ∞. It means, that the described research of dependence of magnetic properties on the shape of the sample is equivalent to the research of demagnetization effects.

The important result of our investigation is that in the sample of spheroidal shape not one but a discrete number of nuclear eigenfrequencies exist. These eigenmodes can be observed in NMR experiment. Of course it is hard to identify index of the observed mode from NMR experiment. But as it was shown, some of the lowest modes cross each other. So, if these crosspoints of eigenmodes are obsered in experiment it may help to identify the index of modes and the type of magnetic precession, that realized in the sample. The discreteness of eigenfrequencies also can produce the specific properties of nonlinear NMR at the low temperatures especially if we deal with a big value of perturbing magnetic field $\mathbf{h}$. Such problem has been investigated in [4]. The main idea of the theory was the conservation of nuclear magnetic moment. The dynamic frequency shift (pulling) depends on the m_z-component of nuclear magnetization and has quite a big value. This pulling also depends on the electronic and nuclear eigenfrequencies. If these eigenfrequencies represent a discret set, the dependence of pulling on the magnetic field H_0 and on m_z-component of nuclear magnetization becomes more complicated. The investigation of this specific pulling is outside of the frames of current paper, but we can predict one result qualitatively. The theoretical NMR dissipation lines in [4] were smooth, while the experimental signal contained additional inhomogeneousnesses, which can be considered as oscillations upon the main line. These oscillations may be described as the effects of discreteness due to the finiteness of the sample and boundary effects.

Acknowledgments

The work was performed within the State contract of the Federal Agency of Scientific Organizations (issue “Spin”, no. AAA-A18-118020290104-2). M.A. Borich acknowledges the support of the Russian sciences foundation, project no. 16-12-10514.

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