Angular dependence of FMR frequency in strongly anisotropic uniaxial ferromagnets

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A diagram technique in the atomic representation is used to calculate the dependence of the FMR frequency on the orientation of the external magnetic field for ferromagnets with strong one-ion anisotropy. The quantum corrections for the FMR frequency are obtained in second order in the parameter $(2S-1)D/(g\mu_B + I_0S)$ at arbitrary values of the spin S. Account is taken of the magnetodipole and anisotropic-exchange interactions. Specific calculations are made for the anisotropic ferromagnet $NiSiF_6 \cdot 6H_2O$, in which quantum effects due to one-ion anisotropy and leading to a change of the H-T diagram were observed in experiment. It is shown that in an external magnetic field $H = 2$ kOe the quantum correction for the FMR frequency of a spherical nickel-fluorosilicate sample reaches $\sim 15\%$ of the phenomenological frequency shift and can be experimentally observed by investigating the dependence of the FMR frequency on the orientation of the external magnetic field.

1. INTRODUCTION

A quantum-theoretic analysis of ferromagnetic resonance (FMR) and of the spectrum of long-wave magnetization oscillations in isotropic ferromagnets leads to the same conclusions as an analysis of the Landau-Lifshitz equation.[1] The situation is more complicated in anisotropic and multisublattice magnets. Thus, for anisotropic ferromagnets, the effective field in the Landau-Lifshitz equation contains an additional anisotropy field. This method of taking the magnetocrystalline anisotropy in account must, of course, be corroborated and has its limitations, as pointed out by Landau.[1)] The concept of the anisotropy field is valid when the characteristic energy of the relativistic interactions is much less than the exchange-interaction energy,[2] and this situation obtains in ferromagnets with relatively high magnetic-ordering temperatures.

As shown by Oguchi,[3] the expression for the frequency of the antiferromagnetic resonance (AFMR) of collinear antiferromagnets contains additional terms of quantum origin. Oguchi calculated the first quantum correction to the AFMR spectrum of uniaxial antiferromagnets. The quantum correction for the AFMR frequency with allowance for anisotropy in the basal plane was obtained by Kukharenko.[4] It was also shown that allowance for the quantum effects explains the experimentally observed nonzero longitudinal susceptibility of transition-metal fluorides as $T \to 0$.

The impossibility of explaining fully the dynamic and static properties of anisotropic magnets on the basis of equations of motion of the Landau-Lifshitz type stimulated the development of model-free theories[5–7] that are applicable in a wide temperature range. This approach has made it possible to take fullest account of the symmetry properties of the system and to determine the relations between the static and dynamic characteristics.

In magnets with strong one-ion anisotropy (OA), the states of the magnetoactive ion constitute, generally speaking, mixtures of states with different components along the quantization axis.[8–12] This leads, in particular, to the appearance of longitudinal and additional transverse branches of the spin-wave excitation spectrum,[8,10–12] and also to a decrease of the saturation magnetization even at zero temperature. These effects, whose intensity increases with increase of the OA, have by now been experimentally observed in intermetallic compounds having a strong OA.[13–18]

The quantum properties of the OA are manifested also by a substantial restructuring of the form of the phase diagrams of anisotropic magnets, when the OA constant is larger than or comparable to the exchange interaction.[19–21] In this context, great interest attached to nickel fluorosilicate $NiSiF_6 \cdot 6H_20$, in which the variation of the magnetic state with change of the OA parameter could be traced.[20]

The spectral characteristics of anisotropic ferromagnets also have distinctive properties of quantum origin.[8,12,22–26] If the OA energy is comparable with the exchange-interaction energy, a spin-wave theory can be constructed with exact allowance for the OA (Refs. 8, 12, 22, 24, and 27–29). When the exchange interaction plays the principal role, the quantum corrections to the properties of the ground state and the spin-wave excitation spectrum can be calculated by expansion in powers of D/I_0 (D is the anisotropy parameter and I_0 the exchange integral).[25,30,31] The quantum corrections are obtained here for arbitrary values of the spin S.

The purpose of the present paper is a calculation of the dependence of the FMR frequency of a uniaxial ferromagnet on the orientation of the external magnetic field, with allowance for quantum effects. It is assumed that the magnetic field is stronger than the anisotropy field. The problem contains therefore, as in Ref. 25, a small parameter $D/(I_0S + g\mu_B H)$ for the expansion. We use in the calculation Matsubara Green's functions calculated by a diagram technique for Hubbard operators.[8] Account is taken of magnetodipole and of anisotropic exchange interactions. The dispersion equation is solved accurate to second order in $D/\overline{H}$, and the quantum correction to the angular dependence of the FMR frequency is obtained in explicit form.

Specific predictions are made concerning the uniaxial ferromagnet $Ni\,SiF_6 \cdot 6H_2O$. It is shown that the FMR frequency shift produced in this substance by quantum effects reaches $\sim 15\%$ of the frequency shift determined by the phenomenological expression.

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2. HAMILTONIAN OF THE PROBLEM

Consider a uniaxial ferromagnet in an external magnetic field H making an angle θ_H with the anisotropy axis. The Hamiltonian of such a system can be written in the form

$$\mathcal{H} = -\frac{1}{2}\sum_{fg}(\mathbf{S}_f \hat{I}_{fg}^{0}\mathbf{S}_g) - \sum_f \{2D_2(S_f^z)^2 + g\mu_B(\mathbf{H}^0\mathbf{S}_f)\} + \frac{g^2\mu_B^2}{2}\sum_{fg}\left\{\frac{(\mathbf{S}_f\mathbf{S}_g)}{R_{fg}^3} - \frac{3(\mathbf{R}_{fg}\mathbf{S}_f)(\mathbf{R}_{fg}\mathbf{S}_g)}{R_{fg}^5}\right\}, \tag{1}$$

where the first term describes the anisotropic exchange interaction, and the second takes into account the uniaxial-symmetry crystal field and the Zeeman-interaction energy. The last term describes the magnetodipole interaction between localized magnetic moments.[32] The zero superscript means that the matrix $\hat{I}_{fg}^0$ and the vector $\mathbf{H}^0$ are written in a coordinate frame in which the z axis coincides in direction with the anisotropy axis. Then, without loss of generality, we can assume that

$$(\hat{I}_{fg}^{0})_{ij} = \delta_{ij} I_{fg}^{i}, \quad I_{fg}^{1} = I_{fg}^{2} \equiv I_{fg}^{\perp}, \quad I_{fg}^{3} \equiv I_{fg}^{\parallel}, \quad \mathbf{H}^0 = (H\sin\theta_H,\ 0,\ H\cos\theta_H). \tag{2}$$

For future convenience, we introduce a three-dimensional matrix $\hat{F}_{fg}^0$ such that the magnetodipole interaction energy operator $\mathcal{H}_{md}$ has the following structure:

$$\mathcal{H}_{md} = -\frac{1}{2}\sum_{fg}(\mathbf{S}_f \hat{F}_{fg}^{0}\mathbf{S}_g). \tag{3}$$

The matrix $\hat{F}_{fg}^0$ is then given by the expression

$$\hat{F}_{fg}^{0} = -\frac{g^2\mu_B^2}{R_{fg}^5}\begin{Vmatrix} (R_{fg}^2 - 3X_{fg}^2) & -3X_{fg}Y_{fg} & -3X_{fg}Z_{fg} \\ -3Y_{fg}X_{fg} & (R_{fg}^2 - 3Y_{fg}^2) & -3Y_{fg}Z_{fg} \\ -3Z_{fg}X_{fg} & -3Z_{fg}Y_{fg} & (R_{fg}^2 - 3Z_{fg}^2) \end{Vmatrix}. \tag{4}$$

In a homogeneous magnetic phase, the spontaneous-magnetization vector $\langle\mathbf{S}\rangle$ makes a certain angle θ with the z axis of the initial coordinate frame, and lies in the ZOX plane. To simplify the calculations, we change to a frame in which the magnetization vector is directed along a new axis z' (the prime will be omitted hereafter). This is done by rotating the coordinate frame through an angle θ around the y axis. The change to the new coordinate system means a unitary transformation of the Hamiltonian operator:

$$\mathcal{H} \to \mathcal{H}' = U\mathcal{H}U^{+}, \quad U = \prod_f \exp(i\theta S_f^y). \tag{5}$$

In the new frame, after separating the self-consistent field (SCF), the Hamiltonian of our system can be written as a sum of the one-ion part and a term that takes the correlation effect of the pair interaction into account:

$$\mathcal{H}' = \sum_f \mathcal{H}_0(f) - \frac{1}{2}\sum_{fg}(\Delta\mathbf{S}_f \hat{A}_{fg}\Delta\mathbf{S}_g), \quad \Delta\mathbf{S}_f = \mathbf{S}_f - \langle\mathbf{S}_f\rangle, \tag{6}$$

where

$$\mathcal{H}_0(f) = {}^1/_2 A_0^{33}\sigma^2 - \langle\mathbf{S}_f\rangle\hat{A}_0\mathbf{S}_f - 2D_2(\tilde{S}_f^z)^2 - g\mu_B(\mathbf{H}\mathbf{S}_f), \quad \tilde{S}_f^z = S_f^z\cos\theta - S_f^x\sin\theta, \quad \sigma \equiv \langle S^z\rangle. \tag{7}$$

The matrix $\hat{A}_0$ is the Fourier transform of the matrix $\hat{A}_{fg}$ at zero quasimomentum, and is determined from the expansion

$$\hat{A}_{fg} = \hat{O}(\hat{I}_{fg}^{0} + \hat{F}_{fg}^{0})\hat{O}^{-1} = \frac{1}{N}\sum_{\mathbf{q}} A_{\mathbf{q}}\exp\{i\mathbf{q}(\mathbf{R}_f - \mathbf{R}_g)\}. \tag{8}$$

The similarity transformation with the aid of the matrix

$$\hat{O} = \begin{Vmatrix} \cos\theta & 0 & -\sin\theta \\ 0 & 1 & 0 \\ \sin\theta & 0 & \cos\theta \end{Vmatrix}$$

reflects the change of the form of the pair interaction on changing to the new coordinate frame. The vector $\mathbf{H}$ is then connected with the vector $\mathbf{H}^0$ by the relation $\mathbf{H} = \hat{O}\mathbf{H}^0$. The calculation of the Fourier transforms for the terms of the magnetodipole interaction is described in detail in Ref. 32. For a sample of finite size, bounded by a second-order surface, we can introduce the demagnetizing-coefficients tensor. The terms of the $\hat{A}_0$ matrix elements corresponding to the dipole-dipole interaction can then be expressed via the demagnetizing fields and the anisotropy of the magnetodipole interaction. This anisotropy will be neglected for two reasons. First, we assume that the magnetodipole-interaction anisotropy is small compared with the OA. Second, the magnetodipole anisotropy has a two-ion character and does not lead to quantum effects in the zeroth order in $1/r_0^3$. Allowance for this anisotropy reduces therefore only to the appearance of an additional term in the complete expression for the effective anisotropy constant (24) contained in ω_{cl}. The contribution to the quantum corrections is then manifested by additional terms proportional to the product of the squared OA constants and magnetodipole anisotropy constants, which are of next order of smallness compared with the principal corrections (see below). Assuming next that the axes of the initial coordinate frame coincide with the principal axes of the surface ellipsoid, we get

$$A_0^{33} = I_0^{\perp}\sin^2\theta + I_0^{\parallel}\cos^2\theta - \frac{4\pi g\mu_B M_0}{S}\left[N_1\sin^2\theta + N_3\cos^2\theta - \frac{1}{3}\right], \tag{9}$$

where $M_0 = g\mu_B S/v_0$ is the saturation magnetization, v_0 is the unit-cell volume, and N_i are the principal components of the demagnetizing-coefficients tensor.

3. DISPERSION EQUATION

To calculate the spectrum of the collective excitations of the Hamiltonian (6) with a nonequidistant spectrum of the one-ion energy levels, we use the diagram-technique formalism for Hubbard operators.[8] We introduce the eigenfunctions of the one-ion Hamiltonian:

$$\mathcal{H}_0(f)|\Psi_n\rangle = E_n|\Psi_n\rangle, \quad n = 1, 2, \ldots, 2S+1, \tag{10}$$

which we use to construct the Hubbard operators

$$X_f^{nm} \equiv |\Psi_n(f)\rangle\langle\Psi_m(f)|. \tag{11}$$

In the new representation, the Hamiltonian (6) takes the form

$$\mathcal{H}=\sum_{f,n}E_n h_{fn}-\frac{1}{2}\sum_{fg}\sum_{\lambda\lambda'}\{\mathbf{c}(\lambda)\hat{a}\hat{A}_{fg}\tilde{a}\mathbf{c}(\lambda')\}\Delta X_f^{\lambda}\cdot\Delta X_g^{\lambda'}, \tag{12}$$

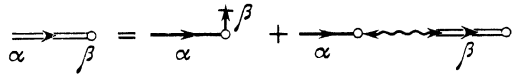
where $h_{fn}\equiv X_f^{nn}$, and the components of the vector $\mathbf{c}(\lambda)$ are parameters of the representation of the spin operators in terms of the Hubbard operators.[29] The matrix $\hat{a}$ effects the transition from the vector $\mathbf{u}_f(S_f^z,\ S_f^+,\ S_f^-)$ to the vector $\mathbf{S}_f=(S_f^x,\ S_f^y,\ S_f^z)$:

$$S_f{}^i=a_{ji}u_f{}^j.$$

The dispersion equation that describes the spectrum of the undamped excitations is determined by the poles of the Fourier transform of the Matsubara Green's function

$$D_{\lambda\lambda'}(f\tau,\ g\tau')=-\langle T\bar{X}_f^{\lambda}(\tau)\bar{X}_g^{\lambda'}(\tau')\rangle,$$

calculated in the zeroth approximation of the SCF method. The graphic series for $D^0_{\alpha\beta}(\mathbf{q},\omega_n)$ is specified by zero-loop diagrams, and the equation for $D^0_{\alpha\beta}(\mathbf{q},\omega_n)$ is[8]



(13)

The solution of this equation is based on the use of the difference between the dependence of the pair-interaction matrix elements on the indices λ and λ' (Ref. 29). After simple calculations we obtain the dispersion equation

$$\det\|1+\hat{L}_0(\omega_n)\hat{V}_{\mathbf{q}}\|=0, \tag{14}$$

where the matrix $\hat{L}_0(\omega_n)$ is defined as

$$\hat{L}_0(\omega_n)=\begin{Vmatrix} u(\omega_n) & v(\omega_n) & v^*(\omega_n)\\ v(-\omega_n) & w(\omega_n) & z(\omega_n)\\ v^*(-\omega_n) & z(-\omega_n) & w(\omega_n)\end{Vmatrix}. \tag{15}$$

The functions $u(\omega_n), v(\omega_n),\ldots$ in (15) can be represented in the low-temperature region ($T\ll T_c$) in the form

$$u(\omega_n)=\sum_{\alpha}\frac{|\gamma_{\parallel}(\alpha)|^2 b(\alpha)}{i\omega_n+\alpha E},$$

$$v(\omega_n)=\sum_{\alpha}\frac{\gamma_{\parallel}^*(\alpha)\gamma_{\perp}(\alpha)b(\alpha)}{-i\omega_n+\alpha E},$$

$$z(\omega_n)=\sum_{\alpha}\frac{|\gamma_{\perp}(\alpha)|^2 b(\alpha)}{i\omega_n+\alpha E},$$

$$w(\omega_n)=\sum_{\alpha}\frac{\gamma_{\perp}(\alpha)\gamma_{\perp}(-\alpha)b(\alpha)}{i\omega_n+\alpha E}. \tag{16}$$

The three-dimensional matrix $\hat{V}_{\mathbf{q}}$ is connected with the matrix $A_{\mathbf{q}}$ of (8) by the relation

$$\hat{V}_{\mathbf{q}}=\hat{a}\hat{A}_{\mathbf{q}}\hat{a},\quad \hat{a}=\frac{1}{2}\begin{Vmatrix}0 & 0 & 2\\ 1 & i & 0\\ 1 & -i & 0\end{Vmatrix}. \tag{17}$$

Equation (14) was obtained under general assumptions concerning the form of the pair interaction and the geometry of the problem. The specific feature of an actual case manifested via the representation parameters $\gamma_{\perp}(\alpha)$ and $\gamma_{\parallel}(\alpha)$ and the one-ion energy levels E_n, and also via the form of the matrix $\hat{V}_{\mathbf{q}}$. In general form, this dispersion equation can hardly be investigated analytically. Examination of various limiting cases, however, enables us to dispense with numerical calculations. This, in particular, is the situation in the homogeneous FMR case, which is of importance in research, subject to the inequalities

$$g\mu_B H\gg 4\pi g\mu_B M_0,\quad (2S-1)|D_2|\gg S|I_0^{\parallel}-I_0^{\perp}|, \tag{18}$$

which allow us to assume that the ferromagnetic sample is in a one-domain state. The specific forms of the representation parameters and of the one-ion energies can then be calculated by perturbation theory.

4. THE ONE-ION PROBLEM

We express the one-ion Hamiltonian in the form

$$\mathcal{H}_0(f)={}^1\!/_2 A_0{}^{33}\sigma^2-\bar{H}_z S_f^z-V_f, \tag{19}$$

where $\bar{H}_z$ is the principal part of the effective field acting on the ion:

$$\bar{H}_z=g\mu_B H\cos(\theta_H-\theta)+A_0{}^{33}\sigma.$$

This field sets the equidistant structure of the unperturbed energy levels:

$$\varepsilon_n={}^1\!/_2 A_0{}^{33}\sigma^2-\bar{H}_z(S-n+1),\quad n=1,2,\ldots,2S+1,$$

and the "bare" states are the eigenstates of the operator S^z. The operator V_f, regarded as a perturbation, is given by

$$V_f=-\bar{H}_x S_f^x-2D_2(S_f^z)^2, \tag{20}$$

where the effective field acting along the x axis is of the form

$$\bar{H}_x=g\mu_B H\sin(\theta_H-\theta)+\frac{1}{2}(I_0^{\perp}-I_0^{\parallel})\sigma\sin 2\theta - g\mu_B M_0 2\pi(N_1-N_3)\frac{\sigma}{S}\sin 2\theta.$$

To separate the quantum effects, second order perturbation theory in V_f is sufficient. We then obtain at low temperatures ($T\ll T_c$)

$$E_n=\varepsilon_n+\langle n|V|n\rangle+E_n^{(2)},$$

$$\sigma=S+\frac{E_1^{(2)}}{\bar{H}_z}=S\left[1-\frac{(2S-1)}{2}\left(\frac{D_2}{\bar{H}_z}\right)^2\sin^4\theta_H\right]. \tag{21}$$

The angle θ in the considered temperature region is obtained from the condition that the energy E_1 be a minimum, and can be written in the form

$$\theta=\theta_0+\delta. \tag{22}$$

The angle θ_0 satisfies the equation

$${}^1\!/_2\bar{D}\sin 2\theta_0=g\mu_B H\sin(\theta_H-\theta_0), \tag{23}$$

which corresponds to the equation obtained in the phenomenological treatment. The effective anisotropy constant

$$\bar{D}=2(2S-1)D_2+S(I_0^{\parallel}-I_0^{\perp})+4\pi g\mu_B M_0(N_1-N_3) \tag{24}$$

takes into account the OA and the anistropy of the exchange

interaction and of the sample shape.

The correction δ in (22) is connected with the OA quantum effects and is of the form

$$\delta=\frac{(2S-1)D_2^2\sin^2\theta_H\sin 2\theta_H}{[g\mu_B H+2\pi g\mu_B M_0(N_1-N_3)\cos 2\theta_H]\overline{H}_z(\sigma=S,\theta=\theta_H)}. \tag{25}$$

Calculating the eigenstates (10) by perturbation theory

$$|\Psi_n\rangle=|n\rangle+|\Psi_n^{(1)}\rangle+|\Psi_n^{(2)}\rangle,$$

we easily obtain, at the accuracy considered, the parameters of the representation

$$\gamma_\perp(\alpha(n,m))=\langle\Psi_n|S^+|\Psi_m\rangle,\quad \gamma_\parallel(\alpha(n,m))=\langle\Psi_n|S^z|\Psi_m\rangle.$$

From Eqs. (16) we get then ($\varepsilon_{nm}=E_n-E_m$, $\overline{H}=\overline{H}_z(\sigma=S,\theta=\theta_H)$)

$$z(\omega_n)=\frac{2S}{i\omega_n-\varepsilon_{21}}\left\{1-\frac{2(2S-1)D_2^2}{\overline{H}^2}\times\left[\sin^2 2\theta-\frac{1}{4}\left(S-\frac{3}{2}\right)\sin^4\theta\right]\right\}$$
$$-\frac{S(2S-1)^2D_2^2}{2(i\omega_n+\varepsilon_{21})\overline{H}^2}\sin^4\theta+\frac{4S(2S-1)D_2^2}{(i\omega_n-\varepsilon_{31})\overline{H}^2}\sin^2 2\theta, \tag{26}$$

$$w(\omega_n)=\frac{2S(2S-1)\varepsilon_{21}}{(i\omega_n)^2-\varepsilon_{21}^2}\times\left[1-2(S-1)(2\cos^2\theta-\sin^2\theta)\frac{D_2}{\overline{H}}\right]\frac{D_2}{\overline{H}}\sin^2\theta, \tag{27}$$

$$v(\omega_n)=\frac{2S(2S-1)\sin 2\theta\sin^2\theta D_2^2}{(i\omega_n+\varepsilon_{21})(i\omega_n+\varepsilon_{31})\overline{H}}, \tag{27'}$$

$$u(\omega_n)=\frac{4S(2S-1)\sin^4\theta}{(i\omega_n)^2-\varepsilon_{31}^2}\frac{D_2^2}{\overline{H}}. \tag{28}$$

It can be seen from (26)–(28) that $z(\omega_n)$ is a quantity of zeroth, $w(\omega_n)$ of first, and $v(\omega_n)$ and $u(\omega_n)$ of second order in the parameter D_2/H. Knowing this, we can write the dispersion equation in simpler form if we restrict ourselves to terms of order not higher than the second in D_2/H.

5. FMR FREQUENCY

The equation describing the lower branch of the spin-wave excitation spectrum takes at our accuracy the form

$$1+V_q^{23}[z(\omega_n)+z(-\omega_n)]+[z(\omega_n)z(-\omega_n)-w^2(\omega_n)]\times[(V_q^{23})^2-(V_q^{22})^2]+2V_q^{12}\{v(\omega_n)+v(-\omega_n)+V_q^{23}[v(\omega_n)z(\omega_n)+v(-\omega_n)z(-\omega_n)]\}=0. \tag{29}$$

The inequalities (18) enabled us to confine ourselves in the derivation of (29) to first-order terms in $4\pi M_0/H$ and of zeroth order in $S|I_0^\perp-I_0^\parallel|/H$ in the expressions that lead to the quantum corrections for the spectrum. We confine ourselves hereafter to the investigation of the frequency of homogeneous FMR. Calculating the Fourier transforms of the matrix elements of the pair interaction at zero quasimomentum, we get

$$V_0^{12}=\frac{1}{4}(I_0^\perp-I_0^\parallel)-\frac{4\pi g\mu_B M_0}{S}(N_1-N_3)\sin 2\theta,$$

$$V_0^{22}=\frac{1}{4}\left\{(I_0^\parallel-I_0^\perp)\sin^2\theta+\frac{4\pi g\mu_B M_0}{S}[(N_2-N_1)\cos^2\theta+(N_2-N_3)\sin^2\theta]\right\},$$

$$V_0^{23}=\frac{1}{4}\left\{I_0^\perp(1+\cos^2\theta)+I_0^\parallel\sin^2\theta+\frac{4\pi g\mu_B M_0}{S}\left[N_3\cos^2\theta-N_1\sin^2\theta-\frac{1}{3}\right]\right\}. \tag{30}$$

Solving Eq. (2) accurate to terms of second order in $D_2/\overline{H}$, we find by making the analytic continuation $i\omega_n\to\omega+i\delta$ that the frequency of he homogenous FMR can be represented in the following form:

$$\omega_0=\omega_{cl}+\Delta\omega_{qu}. \tag{31}$$

Here ω_{cl} describes the usual dependence of the FMR frequency on the orientation of the external magnetic field, which is obtained by the phenomenological approach[33]:

$$\omega_{cl}=\{[g\mu_B H\cos(\theta_H-\theta_0)+\tilde{D}\cos 2\theta_0][g\mu_B H\cos(\theta_H-\theta_0)+\tilde{D}\cos^2\theta_0+4\pi g\mu_B M_0(N_2-N_1)]\}^{1/2},$$

where $\tilde{D}$ is defined by (24).

The quantum correction $\Delta\omega_{cl}$ consists of two terms:

$$\Delta\omega_{qu}=\Delta\omega_1+\Delta\omega_2. \tag{32}$$

The first term is independent of the sample shape of its dependence on the angle θ_H is qualitatively different than that of ω_{cl} (see Fig. 1):

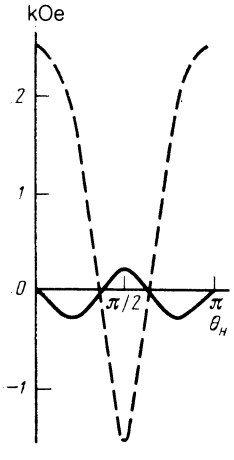


FIG. 1. Angular dependence of phenomenological FMR frequency shift and of the quantum correction for a spherical $NiSiF_6\cdot 6H_2O$ sample in a magnetic field $H=2$ kOe.

$$\Delta\omega_1=\frac{(2S-1)D_2^2}{g\mu_B H+I_0 S}\left[\sin^4\theta_H-\frac{2(1+x)}{1+2x}\sin^2 2\theta_H\right],$$

$$x=I_0 S/g\mu_B H,\quad I_0=(I_0^{\parallel}+I_0^{\perp})/2. \tag{33}$$

The second term of the quantum correction describes the additional FMR frequency shift due to the sample surface shape anisotropy:

$$\Delta\omega_2=-\frac{(2S-1)D_2^2}{2(g\mu_B H+I_0 S)}\left(\frac{4\pi M_0}{H}\right)\left\{\left(\frac{1}{3}-N_3\right)\right.$$

$$\times\left[\frac{7}{(1+x)^2}\sin^4\theta_H-\frac{2(1+x)}{(1+2x)^2}\sin^2 2\theta_H\right]$$

$$+(N_2-N_1)\sin^2\theta_H(2\cos^2\theta_H-\sin^2\theta_H)$$

$$\left.+(N_1-N_3)\sin^2\theta_H\left[\frac{9+5x}{2(1+x)}+\frac{2(1+x)}{(1+2x)^2}\sin^2 2\theta_H\right.\right.$$

$$\left.\left.-\frac{9+2x}{2(1+x)}\sin^4\theta_H\right]\right\}. \tag{34}$$

Quantum corrections to the angular dependence of the FMR frequency are necessitated by the quadrupole moment induced by the OA in the system and by the participation of additional degrees of freedom in the dynamics of the dipole magnetic moment. New modes appear therefore in the collective-excitation spectrum.[8,12,19] Thus, at $\theta_H=\pi/2$ the ferromagnet in question acquires a longitudinal mode due to oscillation of the magnitude of the average magnetization. New transverse modes also appear and are due to collectivization of one-ion excitations corresponding to quadrupole, octupole, etc., transitions. $\theta_H\neq 0$ a distinct classification of the total quantum spectrum into longitudinal and transverse modes is impossible, but is approximately admissible for the investigated weakly anisotropic ferromagnet. The mutual influence of the different spectrum modes is manifested here by an additional shift of the resonant frequencies. Thus, repulsion of the lower spectrum mode from the mode connected with the collectivization of the $1\to 3$ transition leads to a term $\propto\sin^2 2\theta_H$ in the expression for $\Delta\omega_1$. For $\theta_H=\pi/2$ this FMR frequency shift vanishes, for in this geometry the $1\to 3$ shift becomes purely longitudinal and there is no interaction with the transverse mode considered here.

The cause of the contribution to $\Delta\omega_1$ from the term $\propto\sin^4\theta_H$ is the following. Owing to the quantum character of the OA, the average moment at the site decreases at $\theta_H\neq 0$ [see Eq. (21)]. This decrease is manifested by the onset of a corresponding dependence on the one-ion spectrum and the associated spectrum of collective excitations.

The decrease of the magnetization as θ_H increases from 0 to $\pi/2$ via the mechanism of the demagnetizing fields in the presence of the FM surface-shape anisotropy leads also to an additional (on top of the usual phenomenological) shift of the FMR frequency. This effect is described by the correction $\Delta\omega_2$ and depends substantially on the sample shape via the demagnetizing coefficients N_1, N_2, N_3 ($N_1+N_2+N_3=1$).

It follows from the foregoing that the OA quantum effects can be observed by investigating the experimental dependence of the FMR frequency on the angle θ_H in FM with sufficiently large OA.

We examine now the predicted effects for a single-crystal uniaxial ferromagnet—nickel fluorosilicate $NiSiF_6\cdot H_2O$. This single crystal has by now been well investigated and constitutes a model compound with magnetic properties corresponding to the Heisenberg model with uniaxial anisotropy and with spin $S=1$. This compound is also of interest because experiment revealed in it quantum effects due to OA and leading to a unique P-T phase diagram.[20]

At atmospheric pressure and low temperatures, the parameters of the model Heisenberg Hamiltonian that describes well the magnetic properties of $NiSiF_6\cdot 6H_2O$ are

$$D_2=0.0865\,K,\ I_0=0{,}17\,K,\ g=2{,}23. \tag{35}$$

These parameter values are discussed in Ref. 21. In the same reference the authors, using Eq. (35), constructed a theoretical H-T phase diagram that agrees well with the H-T diagram determined for nickel fluorosilicate from the shifts of the specific-heat peaks with change of the magnetic field (see Ref. 21 for the pertinent references).

We shall assume that the $NiSiF_6\cdot 6H_2O$ sample is spherical ($N_1=N_2=N_3=\frac{1}{3}$) and is in an external magnetic field $H=2$ kOe. Using Eqs. (31)–(33) and the parameters (35), it is easy to calculate the dependence of the FMR frequency on θ_H. Figure 1 shows the angular dependences of the classical frequency shift $\omega_{cl}-g\mu_B H$ (dashed line) and of the predicted quantum correction (solid). It is seen that at certain values of θ_H the quantum effects yield an FMR frequency shift amounting to $\sim 15\%$ of the phenomenological shift, and can be experimentally observed in $NiSiF_6\cdot 6H_2O$. It is important that the angular dependence of $\Delta\omega_{qu}$ differs qualitatively from that of ω_{cl}. This circumstance can facilitate considerably the separation of the quantum corrections from the total angular dependence of the FMR frequency when the experimental data are reduced.

Note that the presence of quantum corrections that depend on the sample shape makes it possible to investigate their influence by varying the geometry of the experiment and the type of surface of the investigated ferromagnet.

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[1)]This circumstance was noted by I. E. Dzyaloshinskiĭ in a discussion of the present paper.

[1]L. D. Landau and E. M. Lifshitz, Phyz. Zs. Sowjet. **8**, 153 (1935). L. D. Landau, *Collected Works*, Pergamon, 1965.
[2]L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media*, Pergamon, 1984.
[3]T. Oguchi, Phys. Rev. **117**, 117 (1960).
[4]B. G. Kukharenko, Zh. Eksp. Teor. Fiz. **69**, 632 (1975) [Sov. Phys. JETP **42**, 321 (1975)].
[5]Yu. M. Gufan, *ibid.* **60**, 1537 (1971) [**33**, 831 (1971)].
[6]I. E. Dzyaloshinskiĭ and B. G. Kukharenko, *ibid.* **70**, 2360 (1976) [**43**, 1232 (1976)].
[7]A. F. Andreev and V. I. Marchenko, Usp. Fiz. Nauk **130**, 39 (1980) [Sov. Phys. Usp. **23**, 21 (1980)].
[8]R. O. Zaĭtsev, Zh. Eksp. Teor. Fiz. **68**, 207 (1975) [Sov. Phys. JETP **41**, 100 (1975)].
[9]V. V. Druzhinin, S. P. Zapasskiĭ, and V. M. Povyshev, Fiz. Tverd. Tela (Leningrad) **17**, 23 (1975) [Sov. Phys. Solid State **17**, 12 (1975).
[10]V. M. Loktev, Fiz. Nizk. Temp. **7**, 1184 (1981) [Sov. J. Low Temp. Phys. **7**, 575 (1981)].
[11]B. D. Dunlap, L. M. Hall, F. Behroozi, *et al.* Phys. Rev. **B20**, 6244 (1984)].
[12]F. P. Onufriev, Zh. Eksp. Teor. Fiz. **89**, 2270 (1985) [Sov. Phys. JETP **62**, 1311 (1985)].

[13]K. H. T. Buschow, Rep. Progr. Phys. **42**, 1373 (1979).
[14]A. Furrer and H.-G. Purwins, Phys. Rev. **B16**, 2131 (1977).
[15]W. Schelp, A. Leson, W. Drewes, H.-G. Purwins, and H. Grimm, Z. Phys. **B51**, 41 (1983).
[16]B. Barbara, J. K. Boucherle, M. F. Rossignol, and T. Schweizer, Physica (Utrecht) **B86-89**, 83 (1977).
[17]W. Schelp, W. Drewes, H.-G. Purwins, and G. Eckold, Sol. St. Comm. **52**, 825 (1984). J. Magn. Magn. Mat. **50**, 147 (1985).
[18]B. Frick, M. Loewenhaupt, A. Murani, and R. Feile, *ibid.* **54**, 563 (1985).
[19]E. B. Rozenfel'd, Pis'ma Zh. Eksp. Teor. Fiz. **24**, 60 (1976) [JETP Lett. **24**, 50 (1976)].
[20]V. G. Bar'yakhtar, I. M. Vitebskiĭ, A. A. Galkin, *et al.*, Zh. Eksp. Teor. Fiz. **84** 1083 (1983) [Sov. Phys. JETP **57**, 628 (1983)].
[21]V. G. Borisenko and Yu. V. Preverzev, Fiz. Nizk. Temp. **10**, 946 (1984) [Sov. J. Low Temp. Phys. **10**, 493 (1984)].
[22]I. G. Gochev and V. M. Tsukernik, Fiz. Tverd. Tela (Leningrad) **15**, 1963 (1973) [Sov. Phys. Tech. Phys. **15**, 1315 (1973)].
[23]V. M. Loktev and V. S. Ostrovskii, Ukr. Fiz. Zh. **23**, 1708 (1978).
[24]D. A. Garanin and V. S. Lutovinov, Teor. Mat. Fiz. **55**, 106 (1983).
[25]A. V. Chubukov, Fiz. Nizk. Temp. **10**, 1166 (1984) [Sov. J. Low Temp. Phys. **10**, 609 (1984)].
[26]A. V. Chubukov, J. Phys. **C17**, 1991 (1984).
[27]S. L. Ginzburg, Fiz. Tverd. Tela (Leningrad) **12**, 1805 (1970) [Sov. Phys. Solid State **12**, 1429 (197)].
[28]L. M. Noskova, *ibid.* **18**, 1669 (1976) [**18**, 969 (1976)].
[29]V. V. Val'kov, T. A. Val'kova, and S. G. Ovchinnikov, Zh. Eksp. Teor. Fiz. **88**, 550 (1985) [Sov. Phys. JETP **61**, 323 (1985)].
[30]V. S. Ostrovskiĭ and E. G. Petrov, Phys. Stat. Sol. (b) 41, 369 (1975).
[31]S. P. Obukhov, Fiz. Tverd. Tela (Leningrad) **18**, 1098 (1976) [Sov. Phys. Solid State **18**, 627 (1976)].
[32]A. I. Akhiezer, V. G. Bar'yakhtar, and S. V. Peletminskii, *Spin Waves*, Wiley, 1968.
[33]A. G. Gurevich, Magnetic Resonance in Ferrites and in Antiferromagnets [in Russian], Nauka, 1973.

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