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Derivation of the resonance frequency from the free energy of ferromagnets

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The general form for the magnetic resonance frequency ω of anisotropic ferromagnets as derived from the free energy F by Smit and Beljers $(\omega/\gamma)^2=(M\sin\theta)^{-2}(F_{\theta\theta}F_{\phi\phi}-F_{\theta\phi}^2)$, although numerically correct, is physically not convenient, because the origin of the different terms in F is obscured by an angular-dependent mixing. This mixing is avoided by using the relation

$$\left[\frac{\omega}{\gamma}\right]^2=\frac{1}{M^2}\left[F_{\theta\theta}\left[\frac{F_{\phi\phi}}{\sin^2\theta}+\frac{\cos\theta}{\sin\theta}F_\theta\right]-\left[\frac{F_{\theta\phi}}{\sin\theta}-\frac{\cos\theta}{\sin\theta}\frac{F_\phi}{\sin\theta}\right]^2\right].$$

Explicit expressions will show the symmetry of each of the terms in F for all magnitudes and directions of **H**. In addition, an alternate method which uses only rectangular coordinates and which can easily be generalized for multisublattice systems is described.

I. THE FORM OF SMIT AND BELJERS

Investigating the angular dependence of the ferromagnetic resonance frequency ω is well established[1] for the determination of the anisotropy energy constants in the free energy F, defined as a sum of terms proportional to products of the components M_i of the magnetization, described in a rectangular coordinate system.

Using the classical equation of motion

$$\frac{1}{\gamma}\frac{d\mathbf{M}}{dt}=\mathbf{T}\ , \tag{1}$$

where γ is the gyromagnetic ratio and **T** is the torque acting on the magnetization **M**, it is an elementary procedure to evaluate the resonance frequency ω as a function of F.

Since the first article by Kittel[2] in 1947, several authors[3,4] have gradually increased the complexity of the explicit solutions by weakening the restrictions for their validity.

In 1955, Smit and Beljers (SB) (Ref. 5) treated the general case and published a simple form for ω in terms of second derivatives $\partial^2F/\partial\theta\partial\phi=F_{\theta\phi}$ of the free energy F with respect to the polar angle θ and the azimuthal angle ϕ of the equilibrium magnetization $\mathbf{M}_{eq}$. Except for the singular direction $\theta=0$, where ω may not be obtainable, there are no restrictions for the angles θ and ϕ, in contrast to earlier papers. Their form

$$\left[\frac{\omega}{\gamma}\right]^2=\frac{1}{M^2\sin^2\theta}(F_{\theta\theta}F_{\phi\phi}-F_{\theta\phi}^2) \tag{2}$$

has been published independently by Suhl[6] in 1955 and was found at the same time also by Tannenwald and Lax.[7] This form has also been derived from a Lagrangian formalism by Gilbert,[8] again in 1955. Since then, it is considered to be a standard method.[9–14]

II. MIXED SUBSTRUCTURE OF SB

Why then treat this old subject again? First, it is not convenient that the singular direction $\theta=0$ has to be excluded, since this is a frequently used direction in experiments. Second, and more important, the distinct symmetry properties of the different terms in the free energy F are not visible in the expression of SB for the resonance frequency. The usual assumption that there is a one-to-one correspondence between the terms in the free energy F and their second derivatives in the form of SB is naive and could result in erroneous interpretations. What is the reason for this unusual behavior of SB? Dependent on the orientation of the magnetization, all terms in the free energy F are split into two fractions, except the Zeeman term. Only one fraction corresponds to the second derivative of F and is, therefore, visible in the SB form explicitly. The other fraction cannot be seen directly, because it is part of the second derivative of the Zeeman term in F, the term being proportional to the applied field

H in the SB form. In reality, the term proportional to H in the SB form is composed of the true Zeeman term and fractions originating from the other terms in F. This splitting is present also when the Zeeman term is small relative to the other terms. This splitting is angular dependent, it is absent for some specific angular variations.

Therefore, the explicit terms in SB might have symmetry properties inadequate to the true symmetries of the corresponding terms in F, since part of the terms are not visible. As an example, the second derivative of a term in F with cubic symmetry looses this symmetry property in the form of SB, since a fraction is split and part of the second derivative of the Zeeman term in F.

This splitting is shown in a simple example for a Zeeman term $F^Z(\mathbf{H},\mathbf{M})=-\mathbf{M}\cdot\mathbf{H}$ with axial symmetry and a term

$$F^{\text{cub}}(K,\mathbf{M})=K(m_x^2m_y^2+m_x^2m_z^2+m_y^2m_z^2)$$

with $m_i=M_i/M$, exhibiting cubic symmetry. The second derivatives of the cubic term in the expression for the resonance frequency should reflect the cubic symmetry. This is not always true for the SB form, as demonstrated by comparing two rotations. First, $\mathbf{H}$ is rotated in the (001) plane with angle $\alpha=\phi$ and $\theta=\pi/2$. The resonance conditions derived by SB (Ref. 5) are then given by

$$\left[\frac{\omega}{\gamma}\right]^2=\left[\frac{F^Z_{\theta\theta}}{M}+\frac{2K}{M}[\tfrac{3}{4}+\tfrac{1}{4}\cos(4\alpha)]\right]\times\left[\frac{F^Z_{\phi\phi}}{M}+\frac{2K}{M}\cos(4\alpha)\right], \quad (3)$$

equal to Kittel's result.[2]

Performing a rotation of $\mathbf{H}$ in the (010) plane, one would expect that the resonance frequency is described by the same form, with now $\alpha=\theta$ and $\phi=0$, since these rotations should give the same angular dependence due to the cubic symmetry of the considered free energy F. However, SB give a different form for the rotation in the (010) plane

$$\left[\frac{\omega}{\gamma}\right]^2=\left[\frac{F^Z_{\theta\theta}}{M}+\frac{2K}{M}\cos(4\alpha)\right]\times\left[\frac{F^Z_{\phi\phi}}{M}+\frac{2K}{M}[\tfrac{1}{2}-\tfrac{1}{2}\cos(2\alpha)]\right]. \quad (4)$$

The terms explicitly proportional to K do not reflect the symmetry with respect to the twofold axis at $\alpha=\pi/4$, with deviations gradually increasing to $2K/M$ for $\alpha\to 0$. This deviation is shown in Fig. 1(a) for the term $M^{-1}(\sin^{-2}\theta)F^{\text{cub}}_{\phi\phi}$. Figure 1(b) displays the same term for a rotation in the $(\bar{1}10)$ plane, together with the corresponding correct solution. These deviations are independent of the relative magnitude of the terms, i.e., of $H/(|K|/M)$.

Similar effects are present also for quadratic terms in F, i.e., demagnetization, uniaxial and orthorhombic anisotropy energy. Since only the term $M^{-1}(\sin^{-2}\theta)F_{\phi\phi}$ deviates, it is evident that the transformation to the curved azimuthal coordinate ϕ has to be reconsidered.

III. A FORM WITH VISIBLE SYMMETRY

In order to find a form in which each term reflects its symmetry, the general expression valid for all directions and magnitudes of H was evaluated by the involved method of Keffer and Kittel,[4] see Eq. (A4). The same expression (A4) was obtained when a rectangular method was used, which is described in Appendix B.

When the expression (A4) is compared term by term with the corresponding expression derived according to Eq. (2), the source of the covering of the symmetries can be seen. Indeed, the SB form can be transformed into the form (A5) by not neglecting terms proportional to the first derivatives F_θ (Ref. 15) and F_ϕ, respectively,

$$\left[\frac{\omega}{\gamma}\right]^2=\frac{1}{M^2}\left[F_{\theta\theta}\left[\frac{F_{\phi\phi}}{\sin^2\theta}+\frac{\cos\theta}{\sin\theta}F_\theta\right]-\left[\frac{F_{\theta\phi}}{\sin\theta}-\frac{\cos\theta}{\sin\theta}\frac{F_\phi}{\sin\theta}\right]^2\right]. \quad (5)$$

For $\theta=\pi/2$, this form is exactly the same as the original form of Eq. (2), since then the prefactor $\cos\theta/\sin\theta$ of the neglected terms vanishes. Therefore, it is not surprising that all the explicit examples of SB were published for $\theta=\pi/2$. Also for all other angles, except $\theta=0$, the original SB form is still numerically correct, since at equilibrium the first derivatives F_θ and F_ϕ are zero. However, this form is then no longer physically convenient since the different terms of the free energy F are mixed. The "Zeeman" term in $F_{\phi\phi}/(\sin^2\theta)$ contains also orthorhom-

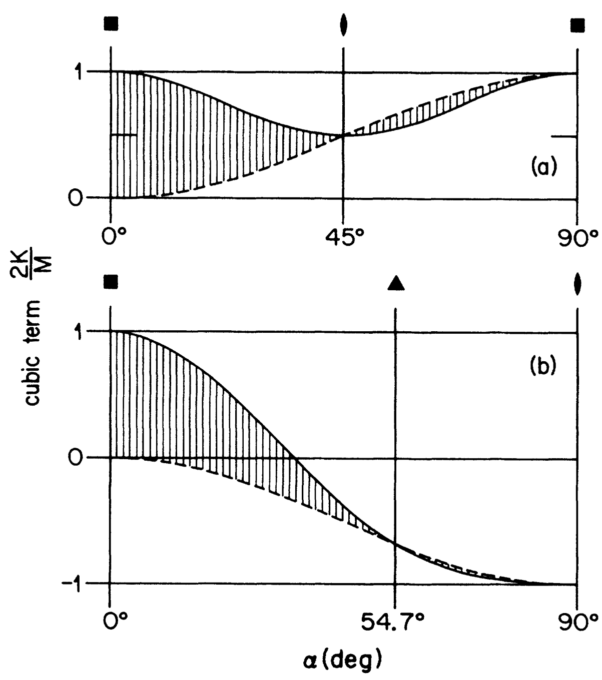

FIG. 1. Angular dependence (broken line) of the cubic anisotropy energy of fourth order in the term $M^{-1}(\sin^{-2}\theta)F^{\text{cub}}_{\phi\phi}$ of Smit and Beljers (Ref. 5), compared to corresponding correct term (solid line), for rotation in (a) (010) plane, (b) $(\bar{1}10)$ plane. The reason for the deviation (shaded) is explained in the text.

bic and cubic terms which are hidden in the neglected term $(\cos\theta/\sin\theta)F_\theta$. Therefore, the important limiting case $\mathbf{M}\|\mathbf{H}$ for a strong field H cannot be found simply by taking the limit $\theta=\theta_H$, $\phi=\phi_H$ for the Zeeman term, because then the orthorhombic and cubic contributions of the neglected terms are lost. The deviations in Fig. 1 are hidden in the Zeeman term of SB. A similar mixing is present also for smaller fields H.

The terms neglected in SB are due to the different curvature of the azimuthal angle ϕ with respect to the rectangular rotational angle η in the form

$$(\omega/\gamma)^2=(1/M^2)(F_{\xi\xi}F_{\eta\eta}-F_{\xi\eta}^2)$$

of SB.[5] Moreover, these neglected terms will make Eq. (5) also valid for $\theta=0$.

The form of Eq. (5) is incorporated in the equations of motion published in 1978.[16] The neglected terms are present in the straightforward evaluation of the derivatives in the curved spherical coordinates for nonequilibrium conditions, thus not neglecting first derivatives.

The new Eq. (5) can also be found using only the original SB form, thus bypassing the geometrical procedure for derivatives of curved coordinates, by assuming small but nonzero deviations $\theta-\theta_H,\phi-\phi_H$ in the Zeeman term of Eq. (2). The resulting terms are replaced by corresponding expressions evaluated from $F_\theta=0$, $F_\phi=0$. Then, the resulting Eq. (5) seems to be defined unambiguously only for large fields H, but there is no mixing for all fields H. Therefore, the original formalism of SB[5] implies also the new form Eq. (5), however, only in an rather implicit fashion.

IV. RECTANGULAR METHOD

The rectangular method of Appendix B combines the advantage of the SB method, i.e., it uses a general solution in a frame with the coordinate ζ along $\mathbf{M}$. Instead of rotational angles ξ,η a rectangular frame denoted $1,2,3\|\zeta$ will be used. Furthermore, the derivatives with respect to angles are replaced by derivatives with respect to the components M_1, M_2, M_3, with the values $M_1=0$, $M_2=0$, and $M_3=M$ at equilibrium. The general expression for the resonance frequency has the form

$$\left[\frac{\omega}{\gamma}\right]^2=(MF_{M_1M_1}-F_{M_3})(MF_{M_2M_2}-F_{M_3})-(MF_{M_1M_2})^2 . \quad (6)$$

This form is equivalent to the form of SB

$$\left[\frac{\omega}{\gamma}\right]^2=\frac{1}{M^2}(F_{\xi\xi}F_{\eta\eta}-F_{\xi\eta}^2) , \quad (7)$$

with $-F_{M_3}$ in Eq. (6) describing the lowering of M_3 when $\mathbf{M}$ is rotated along the rotational angles ξ,η.

The stable equilibrium is found by setting $F_{M_1}=0$ and $F_{M_2}=0$ (with $F_{M_1M_1}>0$ and $F_{M_2M_2}>0$, thus minimizing F), which is equivalent to $M^{-1}F_\theta=0$, $M^{-1}(\sin^{-1}\theta)F_\phi=0$ of SB.

The next task is to transform the free energy F, usually defined in a system x,y,z, which reflects the symmetry of the anisotropy energy into the equilibrium system 1,2,3. Instead of spherical coordinates, an orthogonal transformation $M_i=\sum_\delta b_{i\delta}M_\delta$ is used, defined by a transformation matrix $\tilde{B}$ with Eq. (B8) as an example. After applying this transformation to F, it is then easy to find the relevant terms of the derivatives, and less involved for higher order terms than the SB formalism, as demonstrated in an example for Zeeman interaction and cubic anisotropy energy of sixth order, i.e., for

$$F=-\mathbf{H}\cdot\mathbf{M}+K_{(2)}^{\text{cub}}m_x^2m_y^2m_z^2$$

with $m_i=M_i/M$.

First, the M_i are expressed in the equilibrium components M_δ with the coefficients $b_{i\delta}$ of Eq. (B8),

$$F=-H_x(b_{x1}M_1+b_{x2}M_2+b_{x3}M_3)-H_y(b_{y1}M_1+b_{y2}M_2+b_{y3}M_3)-H_z(b_{z1}M_1+b_{z2}M_2+b_{z3}M_3)$$
$$+\frac{K_{(2)}^{\text{cub}}}{M^6}[(b_{x1}M_1+b_{x2}M_2+b_{x3}M_3)^2(b_{y1}M_1+b_{y2}M_2+b_{y3}M_3)^2(b_{z1}M_1+b_{z2}M_2+b_{z3}M_3)^2] . \quad (8)$$

As an example, the evaluation of the term $MF_{M_2M_2}-F_{M_3}$ in Eq. (6) will be performed. This term corresponds to $M^{-1}F_{\eta\eta}$ of Eq. (7) and is equivalent to the expanded term of SB $M^{-1}(\sin^{-2}\theta)F_{\phi\phi}+(\cos\theta/\sin\theta)F_\theta$ in Eq. (5).

Which terms of F remain after derivation and setting $M_1=0$, $M_2=0$, and $M_3=M$? For F_{M_3}, only terms in F proportional to M_3 and M_3^6 will contribute, yielding 1 and $6M^5$ after derivation. For $F_{M_2M_2}$, the terms proportional to $M_2^2M_3^4$ have to be selected, yielding $2M^4$. Hence the final result is, using $b_{z2}=0$ of Eq. (B8),

$$-F_{M_3}+MF_{M_2M_2}=H_xb_{x3}+H_yb_{y3}+H_zb_{z3}-\frac{K_{(2)}^{\text{cub}}}{M}6b_{x3}^2b_{y3}^2b_{z3}^2+\frac{K_{(2)}^{\text{cub}}}{M}2(b_{x2}^2b_{y3}^2b_{z3}^2+b_{x3}^2b_{y2}^2b_{z3}^2+4b_{x2}b_{x3}b_{y2}b_{y3}b_{z3}^2) . \quad (9)$$

The explicit form is found by inserting the explicit values of the coefficients $b_{i\delta}$ of Eq. (B8).

This method is so simple that the derivation can be performed for all possible terms of a free energy F, see Appendix B. Furthermore, since the general result has the derivation already performed, this method is suitable to computerization and to generalization for multisublattice problems.

V. CONCLUDING REMARKS

The aim of this paper was (i) to show that the form for the resonance frequency of Smit and Beljers[5] is physically not convenient due to a mixing of the different terms in F, and (ii) to propose alternate forms which avoid this mixing. Limiting cases, with a strong field H as an example, are directly visible in these alternate forms.

In addition, explicit expressions are given in Appendix A which do not mix Zeeman, orthorhombic, and cubic anisotropy energy terms, valid for all magnitudes and directions of **H**, including $\theta=0$. Finally, Appendix B describes an alternate method which gives closed forms for all possible terms of the free energy F, including multisublattice systems.

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APPENDIX A

In this appendix equilibrium conditions and the resonance frequency ω are given for the free energy F (Ref. 17)

$$F=-\mathbf{H}\cdot\mathbf{M}+\tfrac{1}{2}(N_x^{\text{eff}}M_x^2+N_y^{\text{eff}}M_y^2+N_z^{\text{eff}}M_z^2)+K_{(1)}^{\text{cub}}(m_x^2m_y^2+m_x^2m_z^2+m_y^2m_z^2)+K_{(2)}^{\text{cub}}m_x^2m_y^2m_z^2\ , \tag{A1}$$

with $m_i=M_i/M$, and with an effective demagnetizing tensor $\tilde{N}^{\text{eff}}$ and cubic anisotropy energy terms $K_{(1)}^{\text{cub}},K_{(2)}^{\text{cub}}$ of fourth and sixth order. Second-order terms $\frac{1}{2}K_im_i^2$ are absorbed in N_i^{eff}.

The equilibrium magnetization $\mathbf{M}_{\text{eq}}$ will be described by the polar angle θ and the azimuthal angle ϕ with respect to the symmetry frame x,y,z. The conditions for equilibrium are

$$\begin{aligned}0=&-H_x\cos\theta\cos\phi-H_y\cos\theta\sin\phi+H_z\sin\theta\\&+(MN_x^{\text{eff}}\cos^2\phi+MN_y^{\text{eff}}\sin^2\phi-MN_z^{\text{eff}})\cos\theta\sin\theta\\&+\frac{2K_{(1)}^{\text{cub}}}{M}[2\cos\theta\sin^3\theta\cos^2\phi\sin^2\phi+\cos\theta\sin\theta(\cos^2\theta-\sin^2\theta)]\\&+\frac{2K_{(2)}^{\text{cub}}}{M}\cos\theta\sin^3\theta\cos^2\phi\sin^2\phi(2\cos^2\theta-\sin^2\theta)\ ,\end{aligned}\tag{A2}$$

$$\begin{aligned}0=&+H_x\sin\phi-H_y\cos\phi+(MN_y^{\text{eff}}-MN_x^{\text{eff}})\sin\theta\cos\phi\sin\phi\\&+\frac{2K_{(1)}^{\text{cub}}}{M}\sin^3\theta\cos\phi\sin\phi(\cos^2\phi-\sin^2\phi)+\frac{2K_{(2)}^{\text{cub}}}{M}\cos^2\theta\sin^3\theta\cos\phi\sin\phi(\cos^2\phi-\sin^2\phi)\ .\end{aligned}\tag{A3}$$

It is, unfortunately, not possible to evaluate the equilibrium angles θ and ϕ from the equilibrium conditions Eqs. (A2) and (A3) for an arbitrary direction of the applied field **H** in closed form. Therefore, approximation techniques[13] are needed and a computer is helpful. However, once these angles are known, the evaluation of the resonance frequency ω is straightforward by using

$$\begin{aligned}\left[\frac{\omega}{\gamma}\right]^2=&\Bigg[H_x\sin\theta\cos\phi+H_y\sin\theta\sin\phi+H_z\cos\theta+(MN_x^{\text{eff}}\cos^2\phi+MN_y^{\text{eff}}\sin^2\phi-MN_z^{\text{eff}})(\cos^2\theta-\sin^2\theta)\\&+\frac{2K_{(1)}^{\text{cub}}}{M}[\cos^4\theta+\sin^4\theta(\cos^4\phi+\sin^4\phi)-3\cos^2\theta\sin^2\theta(1+\cos^4\phi+\sin^4\phi)]\\&+\frac{2K_{(2)}^{\text{cub}}}{M}[\sin^2\theta\cos^2\phi\sin^2\phi(6\cos^4\theta+\sin^4\theta-11\cos^2\theta\sin^2\theta)]\Bigg]\\&\times\Bigg[H_x\sin\theta\cos\phi+H_y\sin\theta\sin\phi+H_z\cos\theta+MN_x^{\text{eff}}(\sin^2\phi-\sin^2\theta\cos^2\phi)\\&+MN_y^{\text{eff}}(\cos^2\phi-\sin^2\theta\sin^2\phi)-MN_z^{\text{eff}}\cos^2\theta\\&+\frac{2K_{(1)}^{\text{cub}}}{M}[\cos^4\theta+\sin^4\theta(\cos^4\phi+\sin^4\phi)-6\sin^2\theta\cos^2\phi\sin^2\phi]\\&+\frac{2K_{(2)}^{\text{cub}}}{M}\{\cos^2\theta\sin^2\theta[\cos^4\phi+\sin^4\phi-(4+3\sin^2\theta)\cos^2\phi\sin^2\phi]\}\Bigg]\end{aligned}$$

$$-\left\{(-MN_x^{\text{eff}}+MN_y^{\text{eff}})\cos\theta\cos\phi\sin\phi+3\frac{2K_{(1)}^{\text{cub}}}{M}[\cos\theta\sin^2\theta\cos\phi\sin\phi(\cos^2\phi-\sin^2\phi)]\right.$$
$$\left.+\frac{2K_{(2)}^{\text{cub}}}{M}[\cos\theta\sin^2\theta(3\cos^2\theta-2\sin^2\theta)\cos\phi\sin\phi(\cos^2\phi-\sin^2\phi)]\right\}^2 . \quad \text{(A4)}$$

APPENDIX B

In this appendix an elementary algebraic method is described which relates the ferromagnetic resonance frequency ω to the free energy F. Similar to Smit and Beljers (SB),[5] a general expression is derived connecting ω to derivatives of F. However, instead of using differentiation with respect to rotational angles, the differentiation will be performed with respect to a rectangular system 1,2,3. This has the advantage that orthogonal transformations can be used, which eliminates the problem of SB[5] involved with the azimuthal angle ϕ.

Suppose that the equilibrium magnetization $\mathbf{M}_{\text{eq}}$ is along 3 of a rectangular coordinate system 1,2,3. In order to evaluate the resonance frequency ω, the torque components T_1, T_2 in

$$\frac{1}{\gamma}\frac{d\mathbf{M}}{dt}=\mathbf{T} \quad \text{(B1)}$$

are expanded as a Taylor series

$$T_j=\sum_{k=1,2}\partial T_j/\partial M_k\,|_{\text{eq}}M_k$$

of the deviations $M_{k=1,2}$ from $\mathbf{M}_{\text{eq}}$. With the notation $A_{jk}=\partial T_j/\partial M_k\,|_{\text{eq}}$, the terms of first order become

$$\frac{1}{\gamma}\frac{dM_1}{dt}=T_1=A_{11}M_1+A_{12}M_2+O(M_1^2,M_2^2)+\cdots , \quad \text{(B2)}$$

$$\frac{1}{\gamma}\frac{dM_2}{dt}=T_2=A_{21}M_1+A_{22}M_2+O(M_1^2,M_2^2)+\cdots . \quad \text{(B3)}$$

The usual procedure of setting $M_j(t)=M_{0j}e^{i\omega t}$ in Eqs. (B2) and (B3) yields the expression for the resonance frequency ω, with the condition $A_{11}=-A_{22}$ for the absence of damping,

$$\left[\frac{\omega}{\gamma}\right]^2=-A_{12}A_{21}-A_{11}^2 . \quad \text{(B4)}$$

Using the relation

$$\mathbf{T}=-\mathbf{M}\times\frac{\partial F}{\partial \mathbf{M}} , \quad \text{(B5)}$$

for a motion which conserves the free energy F and using for brevity

$$F_{M_1}=\partial F/\partial M_1\,|_{M_1=0,M_2=0,M_3=M} ,$$

these coefficients A_{jk} are

$$A_{11}=-A_{22}=MF_{M_2M_1},\quad A_{12}=MF_{M_2M_2}-F_{M_3} ,$$

and

$$A_{21}=-(MF_{M_1M_1}-F_{M_3}) .$$

Thus the resonance frequency ω is connected to the free energy F by the form

$$\left[\frac{\omega}{\gamma}\right]^2=(MF_{M_1M_1}-F_{M_3})(MF_{M_2M_2}-F_{M_3})-(MF_{M_1M_2})^2 . \quad \text{(B6)}$$

The point is that the free energy F is usually defined in the laboratory system x,y,z which reflects the symmetry of the magnetic material, whereas Eq. (B6) is written in the system 1,2,3 with 3 along the equilibrium direction $\mathbf{M}_{\text{eq}}$ defined by $F_{M_1}=0$, $F_{M_2}=0$. Further, it would be convenient to execute the differentiation for a general expression of F

$$F=\sum_{p=0}^{\infty}\sum_{p_x=0}^{p}\sum_{p_y=0}^{p-p_x}C_{p_x,p_y}^{(p)}M_x^{p_x}M_y^{p_y}M_z^{p_z} , \quad \text{(B7)}$$

where the constants $C_{p_x,p_y}^{(p)}$ describe terms of order $p=p_x+p_y+p_z$.

It would be easy to select the relevant terms which survive differentiation and then set $M_1=0$, $M_2=0$, and $M_3=M$ for the equilibrium condition, if F could be expressed in M_1,M_2,M_3.

Assume that the equilibrium direction 3 is known. Then, the orthogonal transformation $\tilde{B}$ with the elements $b_{i,\delta}$ connects both systems

$$\begin{bmatrix}x\\y\\z\end{bmatrix}=\begin{bmatrix}\cos\theta\cos\phi & -\sin\phi & \sin\theta\cos\phi\\ \cos\theta\sin\phi & \cos\phi & \sin\theta\sin\phi\\ -\sin\theta & 0 & \cos\theta\end{bmatrix}\begin{bmatrix}1\\2\\3\end{bmatrix} , \quad \text{(B8)}$$

for 2 within the xy plane with the angle ϕ between the y and 2 axis and θ between z and 3.

Each component M_i in F can be replaced by $M_i=\sum_\delta b_{i\delta}M_\delta$, using the transformation $\tilde{B}$ of Eq. (B8). By doing so, the original constants $C_{p_x,p_y}^{(p)}$ defined in the symmetry system x,y,z remain, but the free energy F is expressed in terms of M_δ with $\delta=1,2,3$.

The general expressions for the derivatives are now easily evaluated as functions of the constants $C_{p_x,p_y}^{(p)}$, the coefficients $b_{i\delta}$, and p,p_x,p_y. The first derivative F_{M_1} is written

$$F_{M_1}=\sum_{p=1}^{\infty}\sum_{p_x=0}^{p}\sum_{p_y=0}^{p-p_x}\sum_{n=x,y,z}^{\text{cycl}} C^{(p)}_{p_x,p_y}M^{p-1}p_n b_{n1}b_{n3}^{p_n-1}b_{(n+1)3}^{p_{(n+1)}}b_{(n+2)3}^{p_{(n+2)}} . \tag{B9}$$

The derivative F_{M_2} is found by replacing 1 with 2. However, the derivative F_{M_3} appearing in Eq. (B6) for the resonance frequency ω is different:

$$F_{M_3}=\sum_{p=1}^{\infty}\sum_{p_x=0}^{p}\sum_{p_y=0}^{p-p_x} C^{(p)}_{p_x,p_y}M^{p-1}p b_{x3}^{p_x}b_{y3}^{p_y}b_{z3}^{p_z} . \tag{B10}$$

For the diagonal second derivatives, only one term is given

$$F_{M_1M_1}=\sum_{p=2}^{\infty}\sum_{p_x=0}^{p}\sum_{p_y=0}^{p-p_x}\sum_{n=x,y,z}^{\text{cycl}} C^{(p)}_{p_x,p_y}M^{p-2} \times[2p_np_{(n+1)}b_{n1}b_{n3}^{p_n-1}b_{(n+1)1}b_{(n+1)3}^{p_{(n+1)}-1}b_{(n+2)3}^{p_{(n+2)}}+p_n(p_n-1)b_{n1}^2b_{n3}^{p_n-2}b_{(n+1)3}^{p_{(n+1)}}b_{(n+2)3}^{p_{(n+2)}}] , \tag{B11}$$

because $F_{M_2M_2}$ is again found by setting $1\rightarrow 2$.

Finally, only one cross term is relevant in Eq. (B6) for the resonance frequency ω:

$$F_{M_1M_2}=\sum_{p=2}^{\infty}\sum_{p_x=0}^{p}\sum_{p_y=0}^{p-p_x}\sum_{n=x,y,z}^{\text{cycl}} C^{(p)}_{p_x,p_y}M^{p-2}[p_np_{(n+1)}(b_{n1}b_{(n+1)2}+b_{n2}b_{(n+1)1})b_{n3}^{p_n-1}b_{(n+1)3}^{p_{(n+1)}-1}b_{(n+2)3}^{p_{(n+2)}} +p_n(p_n-1)b_{n1}b_{n2}b_{n3}^{p_n-2}b_{(n+1)3}^{p_{(n+1)}}b_{(n+2)3}^{p_{(n+2)}}] . \tag{B12}$$

Although the general case seems to be rather involved, the symmetries of the anisotropy energies reduce the number of nonzero constants $C^{(p)}_{p_x,p_y}$ drastically, since they are written in the symmetry system x,y,z. In addition, all terms containing the coefficient b_{z2} vanish, because it is zero for the transformation of Eq. (B8).

The remaining task for a specific free energy F is now reduced merely to the evaluation of the products of the elements $b_{i\delta}$ which connect the system x,y,z to 1,2,3. Moreover, each such product can be written in a simple form

$$\pm\cos^{q_{c\theta}}\theta\,\sin^{q_{s\theta}}\theta\,\cos^{q_{c\phi}}\phi\,\sin^{q_{s\phi}}\phi .$$

Thus for the evaluation of a product only one prefactor and four integers $q_{c\theta}$, $q_{s\theta}$, $q_{c\phi}$, $q_{s\phi}$ are needed. Furthermore, this method can be computerized, since differentiation has been performed already.

[1]For early reviews see, e.g., F. Keffer, in *Encyclopedia of Physics,* edited by S. Flugge and H. P. J. de Wijn (Springer, Berlin, 1966), Vol. XVIII/2, pp. 1–273; E. Kneller, *Ferromagnetismus* (Springer, Berlin, 1962).

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[15]The correction term $(\cos\theta/\sin\theta)F_\theta$ has been introduced by Artman (Ref. 10) for special orientations in order to get the strong-field result equivalent to the Kittel-type formula derived by Blickford (Ref. 3).

[16]P. E. Wigen, in *Physics of Magnetic Garnets,* Proceedings of the International School of Physics, Varenna, Course LXX, edited by A. Paoletti (North-Holland, Amsterdam, 1978), pp. 196–269; see Eq. (8.18) therein.

[17]It might be useful to write the Zeeman term $-\mathbf{H}\cdot\mathbf{M}$ in the form $-HM[\sin\theta\sin\theta_H\cos(\phi-\phi_H)+\cos\theta\cos\theta_H]$, with θ_H,ϕ_H denoting the direction of **H**, see M. Maryško, Czech. J. Phys. B **30**, 1269 (1980); **31**, 1187 (1981); **33**, 686 (1983).