THERMODYNAMIC THEORY OF RESONANCE FREQUENCIES OF ANTIFERROMAGNETS

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The dynamics of spins in antiferromagnets is studied, the usual model assumption regarding conservation of sublattice spin being neglected. It is shown that if dissipation is disregarded certain eigenmodes not considered previously can arise. In antiferromagnets of the easy-axis type the corresponding frequency may have a large, although not completely pure, exchange gap. In other types the new branch may be of the Goldstone type. In ferromagnets the analogous motion is purely dissipative in nature. The third eigenfrequency of antiferromagnets is studied for crystals of the Cr_2O_3 type in the framework of nonequilibrium thermodynamics. Consequences of rejection of the S^2 = const model for the previously studied spectral frequencies are considered. These are the presence of an isotropic gap in the lower branch of the spectrum of antiferromagnets of the α-Fe_2O_3 type and the possibility of hexagonal anisotropy of the spectrum in these crystals in the absence of anisotropy of the static properties. The Goldstone theorem for antiferromagnets is discussed.

IT is well known that relativistic interactions have an important effect on the resonance properties of antiferromagnets. In particular, if anisotropy is disregarded, the gap in the spin wave spectrum of antiferromagnets should be proportional to the magnitude of the external field and not depend on its direction. The large gap in the spectrum of real antiferromagnets finds explanation only if relativistic interactions, which lead to anisotropy, are taken into account[1]. However, a straightforward quantum-mechanical calculation of the resonance frequencies with anisotropy at finite temperatures encounters insurmountable obstacles. Usually for calculations one uses the quasi-classical Landau-Lifshitz equations[2,3], which, besides the energies, have motion integrals associated with the hypothesis that the magnitude of the spin of the sublattices is conserved when inclined from the equilibrium position. This model is quite correct for the description of the oscillations of sublattice spin at low temperatures if each sublattice in an elementary cell can be placed in correspondence with one magnetic atom. This is often not the case. And even if it were, then close to the transition the exchange terms of the thermodynamic potential, which determine the magnitude of the sublattice magnetization, become of the same order as the terms which determine the direction of the magnetic moments,[1)] and by the conservation of motion a deviation from equilibrium must affect the magnitude of the sublattice magnetization. Consideration of these "new" degrees of freedom actually corresponds to taking oscillations of the magnitude of the ordering parameter into account, or else the temperature of the spin subsystem, and of course requires application of the methods of nonequilibrium thermodynamics.

The problem becomes particularly acute for systems which in the usual model are described by several sublattices. The general linear equations of motion have the form

$$\dot{x}_{in} = \gamma_{in,\,kn'}\partial\Delta\Phi / \partial x_{kn'}, \tag{1}$$

where x_{in} is the deviation of the i-th projection of the n-th magnetic sublattice from equilibrium,

$$\Delta\Phi = \alpha_{in,\,kn'}x_{in}x_{kn'} \tag{2}$$

is the addition to the thermodynamic potential of the substance due to the deviation from equilibrium, $\gamma_{in,kn'}$ are phenomenological coefficients, which in the case of dissipationless motion (and we shall consider only this case) obey the relation $\gamma_{in,kn'} = -\gamma_{kn',in}$ (together with the Onsager symmetry rule[4] this makes $\gamma_{in,kn'}$ depend only on odd powers of x^0_{in}). Hence in the case of a single sublattice, because of the antisymmetry of γ_{ik} the dispersion equation for the eigenfrequencies always has a zero solution, i.e., some linear combination of x_i is conserved in the motion.

In the case of the linearized Landau-Lifshitz equations this quantity was the scalar product of the spin deviation $\mathbf{s}$ and its equilibrium value $\mathbf{S}_0$. In the general thermodynamically acceptable equations even for a two-axis ferromagnet in the absence of an external field, it is easy to show by the methods developed below that this is again $\mathbf{s}\cdot\mathbf{S}_0$. It is quite different when $\mathbf{H}_0 \neq 0$. But since resonance experiments consist in determining the coefficients of the series $\omega^2 = a_0 + a_1H_0 + a_2H_0^2 + \ldots$, allowing for anisotropy of the g factor in the Landau-Lifshitz equations[3] gives the same results as the general linear equations of motion. However, for a larger quantity of sublattices the number of such integrals of motion in the general thermodynamically allowable equations of motion does not increase because the effect of forces on one sublattice due to the inclination of another is taken into account.

Thus, in the case of two sublattices (n = 1, 2), the thermodynamic equations of motion are determined by an antisymmetric matrix of the coefficients of the sixth rank. Hence the dispersion equation for the eigenfrequencies, generally speaking, is bi-cubic, of general form with a free term. And the magnetic subsystem in the case of two sublattices must have three

[1)] In ferromagnetics this is manifested by a marked increase in the susceptibility of the paraprocess.

eigenfrequencies of the same kind of resonance. This qualitative result is quite different from the answer obtained from the Landau-Lifshitz equations and some justification is required to use them even in this simplest case.

1. The main features of the new branch of the spectrum of elementary excitations of an antiferromagnet are easily illustrated in the simplest example, when the equilibrium direction of the antiferromagnetism vector $\mathbf{L}$ (taken along z) is given and the anisotropy in the basal plane can be neglected. Consider an antiferromagnetic structure that is even[3] relative to inversion. The magnetic symmetry group in this case is $D_{\infty h}$, and we suppose that the coordinate dependence of $\mathbf{L}(\mathbf{r})$ is not invariant relative to U_2. The thermodynamic equations of motion allowed by the symmetry are

$$l_z = \dot{m}_z = 0, \quad l_x = \beta F_y, \quad l_y = -\beta F_x, \qquad \dot{m}_y = -\beta R_x, \quad \dot{m}_x = \beta R_y, \tag{3}$$

where $\mathbf{m} = \mathbf{x}_1 + \mathbf{x}_2$ and $\mathbf{l} = \mathbf{x}_1 - \mathbf{x}_2$ coincide with the Landau-Lifshitz equations when $\beta = gL_0$.

Here $F_i = \partial\Delta\Phi/\partial m_i$ and $R_i = \partial\Delta\Phi/\partial l_i$ are thermodynamic forces associated with $\mathbf{m}$ and $\mathbf{l}$. The positively defined addition to the thermodynamic potential bilinear in m_i and l_i is

$$\Delta\Phi = {}^1/_2A_1(l_x^2 + l_y^2) + {}^1/_2A_2l_z^2 + {}^1/_2B_1(m_x^2 + m_y^2) + {}^1/_2B_2m_z^2.$$

If we turn on a magnetic field along the antiferromagnetism axis, the magnetic symmetry group becomes $C_{\infty i}$ and the allowed equations of motion are greatly complicated:

$$l_x = \gamma_1R_y + \gamma_2F_x + \beta'F_y, \quad l_y = -\gamma_1R_x - \beta'F_x + \gamma_2F_y, \quad l_z = -\gamma_4F_z, \tag{4}$$

$$\dot{m}_x = -\gamma_2R_x + \beta'R_y + \gamma_3F_y, \quad \dot{m}_y = -\beta'R_x - \gamma_2R_y - \gamma_3F_z, \quad \dot{m}_z = \gamma_4R_z,$$

whereby

$$\Delta\Phi = {}^1/_2A_1'(l_x^2 + l_y^2) + {}^1/_2A_2'l_z^2 + {}^1/_2B_1'(m_x^2 + m_y^2) + {}^1/_2B_2'm_z^2 + d_1(l_xm_y - l_ym_x) + d_2l_zm_z.$$

The phenomenological constants γ_i, as will be seen later, are proportional to the external field, and d_i is the square of the external field. The primes on β, A_i, and B_i mean that these quantities to the accuracy of the applied field coincide with the corresponding constants in (3). In the Landau-Lifshitz equations we have $\gamma_2 = \gamma_4 = 0$, $\beta' = \beta$, $\gamma_i = \gamma_3 = \kappa_{\|0}gH_0$, where $\kappa_{\|0}$ is the parallel susceptibility of the antiferromagnet. The new eigenfrequency $\omega^2 = \gamma_4^2(A_2'B_2' - d_2^2)$ is actually proportional to the square of the external field and is excited by parallel pumping.

With such a simplified approach it is difficult to connect the phenomenological constants γ_4, A_2', B_2', and d_2 with any kind of static measurement and to estimate the order of the excited frequency. Hence for investigations of the new frequency and the other consequences of possible deviations from the model description of spin wave theory we shall carry out the treatment in the framework of nonequilibrium thermodynamics, using the well-known fact that the left sides of (1) transform under symmetry transformations.

2. To avoid any kind of model ideas, we shall use Dzyaloshinskiĭ's concept of spin density[5].[2)] In other words, we shall assume that that part of the true microscopic spin density $\mathbf{S}_N(\mathbf{r})$, the coordinate dependence of which under symmetry operations (appearing in the crystallographic group G of the magnetic cell) is transformed by the α-th row of the n-th irreducible representation of G, which is characterized by unit direction $\mathbf{L}_{\alpha n}(\mathbf{N})$. Here $\mathbf{N}$ is the cell number and $\mathbf{r}$ is a coordinate within it, $\mathbf{L}_{\alpha n}(\mathbf{N})$ are the spin densities (specifically $\mathbf{L}_{A_1g}(\mathbf{N}) = \mathbf{M}(\mathbf{N})$ is the ferromagnetic spin density or magnetic moment of the elementary cell).

Under the action of symmetry operations remaining from the magnetic group, if the time inversion operator T is taken to be the identity, the vectors $\mathbf{L}_{\alpha n}(\mathbf{N})$ with the same n are transformed into each other as the corresponding rows multiplied by a pseudovector. If (leaving out the vector index) the set $\{\mathbf{L}_{\alpha n}\}$ makes up the regular representation of the indicated group, the determination of the equilibrium values $\mathbf{L}^0_{\alpha n}$ as a function of external field $\mathbf{H}_0$ and temperature gives complete information about the static behavior of the magnetic subsystem. In examining the dynamic behavior it is necessary to consider that the eigenfrequencies can to a marked degree by determined by the oscillations of $\mathbf{L}_{\alpha n}(\mathbf{N})$, the average equilibrium values of which are equal to zero. Moreover, in deviating from equilibrium the spin densities may not behave as a single whole. (Of course, in the latter case the very concept of spin density loses meaning, and model representations are needed to describe such excitations.) But if we are interested only in the lower frequencies, we can make the following assertions on the basis of nonequilibrium thermodynamics.

Let the equilibrium configuration of the magnetic subsystem in some relativistic approximation (or model)[5] be described by k spin densities $\mathbf{L}_{\alpha_1 n_1}$, $\mathbf{L}_{\alpha_2 n_2}, \ldots, \mathbf{L}_{\alpha_k n_k}$. Place in correspondence to the set $\{\mathbf{L}_{\alpha_1 n_1}\}$ the set of orthonormal functions $\varphi_{\alpha_1 n_1}$, $\varphi_{\alpha_2 n_2}, \ldots, \varphi_{\alpha_k n_k}$, each of which transforms according to the α_i-th row of the n_k irreducible representation of G. Then for a description of the dynamic behavior to the same degree of accuracy it is necessary to use m spin densities for which the corresponding set of functions $\varphi_{\alpha_1 n_1}, \ldots, \varphi_{\alpha_k n_k}, \ldots, \varphi_{\alpha_m n_m}$ form a group i.e., any pair of them form a product that transforms like one of the functions of this set.

A proof based on the fact that among the coefficients $\gamma_{in,kn'}$ describing the effect of the deviation of a certain spin density from equilibrium on the motion of the others one can find large ones—the exchange coefficients, independent of crystal direction, and small ones—relativistic, is given in[7].

It follows from this, in particular, that in describing the dynamics of any magnetically ordered substance it is necessary to take into account the ferromagnetic spin density. Only in the simplest case, when the coordinate dependence of the basic antiferromagnetic

spin density makes a one-dimensional representation of G, can we consider only the two spin densities **L** and **M** in describing the dynamics. We shall build the entire treatment on this simplest case.

One more assertion[7] is necessary for what follows. If we expand the thermodynamic potential Φ in any small parameter (e.g., in the gradients of the spin densities), then to maintain accuracy in description of the dynamics we must expand it to same degree as we do γ_{ik}. And if the expansion is in the spin densities, then without exceeding the accuracy in the description of the dynamics we can expand γ_{ik} to one degree less. However, close to the point where the magnetic structure changes all terms of the expansion of Φ become of the same order, and it is impossible to expand Φ in the spin densities. For the coefficients of the equations of motions there is no such special temperature interval, and terms of the third order are always less than terms of the first (if these are not forbidden by symmetry). Hence the approximation used below for concrete calculations in which $\gamma_{in,kn'}$ are expanded only to the first power of the spin densities, whereas for Φ the entire series is kept, makes sense.

3. Let us write out the general allowed exchange equations of motion, linear in the forces that arise with deviation from the equilibrium position, if a single wave with a given wave vector **q** propagates in the antiferromagnetic crystal. We take the coefficients of the equations of motion to be whole rational functions of the projections of the equilibrium spin densities antiferromagnetic **L** and ferromagnetic **M**. We take the vector **q** as small enough that all formulas retain terms no higher in order than q^2. The exchange symmetry group is the space group K_h supplemented by the time inversion operator T. The defining elements of K_h are E, I, and σ_h[3]. To take in all types of antiferromagnets which can be described by two spin densities, we consider further the operation of τ-translation by a crystallographic period in any direction.

We divide antiferromagnets into three types. Vectorial: **L** changes sign under I and σ_h (as **q**). Weak ferromagnets: **L** changes sign under σ_h. Pure antiferromagnet with doubling of period: **L** changes or doesn't change sign under I and σ_h, but changes sign under τ.

For the vectorial antiferromagnet in the general case the equations of motion have the form

$$\mathbf{m} = [\mathbf{A}_1\mathbf{F}] + [\mathbf{B}_1\mathbf{R}] + \mathbf{B}_2(\mathbf{R}), \quad \dot{\mathbf{l}} = [\mathbf{B}_1\mathbf{F}] + [\mathbf{A}_2\mathbf{R}] - \mathbf{B}_2(\mathbf{F}), \qquad (5)^*$$

where

$$\mathbf{A}_1 = a_1\mathbf{M}_0 + a_2(\mathbf{L}_0\mathbf{M}_0)\mathbf{L}_0 + a_3(\mathbf{q}\mathbf{M}_0)\mathbf{q} + a_4(\mathbf{L}_0\mathbf{M}_0)(\mathbf{q}\mathbf{L}_0)\mathbf{q} + a_5[\mathbf{q}\mathbf{L}_0] + a_6(\mathbf{L}_0\mathbf{M}_0)[\mathbf{q}\mathbf{M}_0],$$
$$\mathbf{B}_1 = b_1\mathbf{L}_0 + b_2(\mathbf{L}_0\mathbf{M}_0)\mathbf{M}_0 + b_3(\mathbf{q}\mathbf{L}_0)\mathbf{q} + b_4(\mathbf{L}_0\mathbf{M}_0)(\mathbf{q}\mathbf{M}_0)\mathbf{q} + b_5[\mathbf{q}\mathbf{M}_0] + b_6(\mathbf{L}_0\mathbf{M}_0)[\mathbf{q}\mathbf{L}_0];$$

$\mathbf{A}_2$ coincides in form with $\mathbf{A}_1$, but has a_i' instead of a_i; $\mathbf{B}_2$ is a linear vector function with vectorial argument:

$$\mathbf{B}_2(\mathbf{c}) = b_7\mathbf{M}_0([\mathbf{L}_0\mathbf{M}_0]\mathbf{c}) + b_9(\mathbf{M}_0\mathbf{c})[\mathbf{L}_0\mathbf{M}_0] + b_{11}[\mathbf{L}_0\mathbf{q}](\mathbf{q}\mathbf{c}) + b_{13}([\mathbf{L}_0\mathbf{q}]\mathbf{c})\mathbf{q} + b_{15}[\mathbf{L}_0\mathbf{q}]([\mathbf{L}_0\mathbf{M}_0]\mathbf{c}) + b_{17}([\mathbf{L}_0\mathbf{q}]\mathbf{c})[\mathbf{L}_0\mathbf{M}_0] + b_{19}(\mathbf{M}_0\mathbf{q})\mathbf{c} + b_{21}(\mathbf{M}_0\mathbf{c})\mathbf{q} + \mathbf{Q};$$

Q represents terms obtained from those written by replacing $b_i\mathbf{L}_0$ by $b_{i+1}(\mathbf{L}_0\cdot\mathbf{M}_0)\mathbf{M}_0$ and $b_i\mathbf{M}_0$ by $b_{i+1}(\mathbf{L}_0\cdot\mathbf{M}_0)\mathbf{L}_0$; a_i and b_i are whole rational functions of $\mathbf{L}_0^2$, $\mathbf{M}_0^2$, $(\mathbf{L}_0\mathbf{M}_0)^2$, $(\mathbf{q}\mathbf{L}_0)^2$, $(\mathbf{q}\mathbf{M}_0)^2$, $\mathbf{q}^2$, and $(\mathbf{q}[\mathbf{L}_0\times\mathbf{M}_0])$.

For the other two types of antiferromagnet, the equations of motion take the same form, but $a_5 = a_6 = b_5 = b_6 = b_{15} = b_{17} = b_{19} = b_{21} = 0$, and the last argument of the scalar functions a_i and b_i disappears.

Equations (5) permit, if equilibrium $\mathbf{L}_0$ and $\mathbf{M}_0$ are known, the calculation of the exchange terms γ_{ik}, ρ_{ik}, and λ_{ik} in the general thermodynamic equations (the tilde signifies transportion):

$$\dot{m}_i = \gamma_{ik}F_k + \lambda_{ik}R_k, \quad \dot{l}_i = -\tilde{\lambda}_{ik}F_k + \rho_{ik}R_k. \qquad (6)$$

In this general form we can write the criterion for the absence of the third frequency in the system. The secular equation for (6) has the form

$$\omega^6 - \omega^4\,\mathrm{Sp}\,K_1 + \omega^2\,\mathrm{Sp}\,K_2 - \Delta_1\Delta_2 = 0, \qquad (7)$$

where K_1 and K_2 are matrices given in general form in[7], $\Delta_1 = |\alpha_{ik}|$ is the determinant of the matrix of the coefficients of the addition to the thermodynamic potential

$$\Delta_2 = \{\gamma_1\rho_3\lambda_7 - \gamma_1\rho_2\lambda_8 + \gamma_1\rho_1\lambda_9 - \gamma_2\rho_3\lambda_4 + \gamma_2\rho_2\lambda_5 - \gamma_2\rho_1\lambda_6 + \gamma_3\rho_3\lambda_1 - \gamma_3\rho_2\lambda_2 + \gamma_3\rho_1\lambda_3 - |\lambda_{ik}|\}^2, \qquad (8)$$

where $|\lambda_{ik}|$ is the determinant of the matrix $||\lambda_{ik}||$ and the subscripts correspond with numeration in the matrices γ, ρ, λ from left to right and top to bottom.

If in the general equations of motion we go over to sublattice notation, the form of Δ_2 does not change and we see at once that if the forces arising in deviation from equilibrium position in one sublattice do not affect the motion of the other ($\lambda_{ik} = 0$), then Δ_2, and with it the new resonance frequency, goes to zero. This also comes out of the Landau-Lifshitz equations.

4. One more general result—the Goldstone theory for antiferromagnets—is easy to obtain if we transform to spherical coordinates in ϵ-space[8]. This can be formulated in the following way[9].

If the ground state of the antiferromagnet is degenerate with respect to any coordinate, then in the spectrum there will be a collective branch with frequency that goes to zero together with **q**. The normal coordinates corresponding to this frequency are related linearly to the coordinate with respect to which the state is degenerate.

Consider a uniaxial antiferromagnet with symmetry axis C_φ. Besides the fixed xyz coordinate system, we introduce a system x'y'z'. We have $\mathbf{y}' \parallel \mathbf{L}_0$ and $\mathbf{x}'$ parallel to the projection of **M** on the plane perpendicular to **L**. The symbols φ and θ stand for the azimuthal and vertical angles that determine the direction of **L** in the xyz system, and χ and γ the corresponding angles for **M** in x'y'z'. Let $|\mathbf{L}| = S\sin\xi$, $|\mathbf{M}| = S\cos\xi$; then

$$L_x = S\sin\xi\sin\theta\cos\varphi, \quad L_y = S\sin\xi\sin\theta\sin\varphi, \quad L_z = S\sin\xi\cos\theta;$$
$$M_x = S\cos\xi\{-\sin\chi(\cos\gamma\cos\theta\cos\varphi - \sin\gamma\sin\varphi) + \cos\chi\sin\theta\cos\varphi\},$$
$$M_y = S\cos\xi\{-\sin\chi(\cos\gamma\cos\theta\sin\varphi + \sin\gamma\cos\varphi) + \cos\chi\sin\theta\sin\varphi\},$$
$$M_z = S\cos\xi\{\sin\chi\sin\theta\cos\gamma + \cos\chi\cos\theta\}.$$

If we are considering a weak ferromagnet, then for uniform magnetization Φ is an arbitrary function of the following seven invariants, which comprise the whole rational basis of the invariants:

*$[\mathbf{A},\mathbf{F}] \equiv \mathbf{A}\times\mathbf{F}$.

$$I_1 = \mathbf{L}^2, \quad I_2 = \mathbf{M}^2, \quad I_3 = (\mathbf{LM})^2, \quad I_4 = L_z^2,$$
$$I_5 = M_z^2, \quad I_6 = L_z M_z(\mathbf{LM}), \quad I_7 = [\mathbf{LM}]\mathbf{n}.$$

Below, in addition to this system of basis invariants, we use a basis in the coordinates of (9):

$$i_1 = S^2, \quad i_2 = \cos^2\xi, \quad i_3 = \cos^2\chi, \quad i_4 = \cos^2\theta, \quad i_5 = \cos^2\gamma,$$
$$i_6 = \sin 2\chi \sin 2\theta \cos\gamma, \quad i_7 = \sin 2\xi \sin\chi \sin\theta \sin\gamma.$$

In the other antiferromagnets the Dzyaloshinskiĭ invariant $I_7(i_7)$ disappears.

The derivatives of the thermodynamic potential with respect to the first system of functions are symbolized below by roman numerals, with respect to the second by arabic numerals, e.g.,

$$\partial^2\Phi / \partial I_3 \partial I_2 = \Phi_{\mathrm{III,\,II}}, \qquad \partial^2\Phi / \partial i_1 \partial i_4 = \Phi_{14}.$$

If we rewrite the equations of motion in the coordinates (9), then (7) keeps its form, which is associated only with the antisymmetry of the coefficients of the equations of motion. Since $\Delta\Phi$ for a uniform deviation of the magnetic subsystem from equilibrium in this case does not depend on φ, then $\Delta_1 = 0$. And $\omega(q)$ is of interest, by considering the solitary wave with the specified q, the axial symmetry of $\Delta\Phi$ disappears: α_{ik} depends essentially on the mutual orientation of $\mathbf{L}_0$ and q (the orientation of $\mathbf{L}_0$ and $\mathbf{M}_0$ is determined by internal properties of the system), e.g., on account of invariants of the form $(q\cdot L)^2$, $[[q\times L]\times[q\times M]]n$. Thus, the determinant Δ_1, and with it one of the resonance frequencies, goes to zero along with q. A dependence of $\Delta\Phi$ on φ also arises in the presence of an external field $\mathbf{H}_0$. However, the above proof does not exist if the equilibrium value $\sin\theta_0 = 0$. In this case it is impossible to introduce a coordinate φ. The relation between the Goldstone coordinates and the derivations of the introduced coordinates from equilibrium values is better seen in specific examples.

5. Let us consider the deviation from equilibrium of the magnetic subsystem in a weak ferromagnetic with the aid of the exchange equations of motion.

In the absence of an applied magnetic field the equations determining the equilibrium state of the antiferromagnet have the form

$$\begin{aligned} &2S\Phi_1 = 0; \quad -\sin 2\xi\Phi_2 + 2\cos 2\xi \sin\theta \sin\chi \sin\gamma\Phi_7 = 0,\\ &-\sin 2\chi\Phi_3 + 2\cos 2\chi \sin 2\theta \cos\gamma\Phi_6 + \sin 2\xi \sin\theta \cos\chi \sin\gamma\Phi_7 = 0,\\ &-\sin 2\theta\Phi_4 + 2\sin\chi \cos 2\theta \cos\gamma\Phi_6 + \sin 2\xi \cos\theta \sin\chi \sin\gamma\Phi_7 = 0,\\ &-\sin 2\gamma\Phi_5 - 2\sin 2\chi \sin 2\theta \sin\gamma\Phi_6 + \sin 2\xi \sin\theta \sin\chi \cos\gamma\Phi_7 = 0. \end{aligned} \tag{10}$$

Equation (10) has two solutions of Type I in the classification used in[8]:

1) $\sin\chi_0 = \cos\xi_0 = \sin 2\theta_0 = \sin 2\gamma_0 = \Phi_1 = 0;$

2) $\cos\chi_0 = \sin\theta_0 = \cos\xi_0 = \sin 2\gamma_0 = \Phi_1 = 0.$

Anisotropic exchange, described by Moriya[10], is the physical reason for the realization of a solution of (10) of Type II in the same classification:

$$\cos\chi_0 = \cos\theta_0 = \cos\gamma_0 = \Phi_1 = -\sin 2\xi\Phi_1 + 2\cos 2\xi\Phi_7 = 0.$$

We shall be interested only in the latter case, which is realized in α-Fe_2O_3. If $\mathbf{H}_0$ lies in the basal plane, this state is conserved, except it is necessary to take into account that $\mathbf{M}_0 \parallel \mathbf{H}_0$. For uniform motion, if in $\mathbf{A}_i$ and $\mathbf{B}_i$ only terms linear in the equilibrium spin densities remain, system (5) is greatly simplified:

$$\dot{s} = -f_\theta(b_1 - a_1')\cos\xi_0, \quad S_0\dot{\xi} = f_\theta(b_1 + a_1'\operatorname{ctg}^2\xi_0), \tag{11}$$

$$\begin{aligned} &\dot{\theta} = f_s(b_1 - a_1')\cos\xi_0 - \frac{1}{S_0}(b_1 + a_1'\operatorname{ctg}^2\xi_0)f_\xi;\\ &S_0\cos\xi_0\dot{\chi} = -f_\gamma(a_1 - b_1), \quad S_0\cos\xi_0\dot{\varphi} = b_1 f_\gamma,\\ &S_0\cos\xi_0\dot{\gamma} = f_\chi(a_1 - b_1) - f_\varphi b_1. \end{aligned} \tag{12}$$

If we compare this with[11] it is obvious that (12) corresponds to a lower resonance frequency. Since this is precisely the one studied in experiments, we find, considering the relation between I_k and i_1 and the fact that $f_\gamma(\mathbf{H}_0 = 0) = (2\Phi_5 - \sin 2\xi\Phi_7)\Delta\gamma$, and $f_\chi(H_0 = 0) = (2\Phi_3 - \sin 2\xi\Phi_7)\Delta\chi$, that the expansion of the lower resonant frequency in powers of H_0 begins with the free term

$$\Omega^2(\mathbf{H}_0 = 0) = (a_1 - b_1)^2 L_0^2 \Phi_{\mathrm{VII}}^2 \left(1 + \frac{\Phi_{\mathrm{V}}}{\Phi_{\mathrm{II}} - \Phi_{\mathrm{I}}}\right)\left(1 + \frac{\Phi_{\mathrm{III}} L_0^2}{\Phi_{\mathrm{II}} - \Phi_{\mathrm{I}}}\right).$$

To explain the presence of this isotropic gap in the spectrum of α-Fe_2O_3 with the Landau-Lifshitz equations it is necessary to consider magnetoelastic interactions[12,13]. As regards the Goldstone coordinate (the two of them), in this case they are linear combinations of all six variables. The Goldstone frequency ω^2 is proportional to $b_i b_k q^4$ (where i, 1 = 11–14).

Another result that differs from answers obtained from and Landau-Lifshitz equations is that the zz component of the high-frequency susceptibility tensor does not go to zero with H_0:

$$\varkappa_{zz} = \frac{a_1 H_0(\Phi_{\mathrm{VII}} L_0/(\Phi_{\mathrm{II}} - \Phi_{\mathrm{I}}) + \varkappa_0 H_0)}{\omega^2 - \Omega^2(H_0)} + \frac{(a_1 - b_1)^2 L_0^2 \Phi_{\mathrm{VII}}^2}{2(\Phi_{\mathrm{II}} - \Phi_{\mathrm{I}})[\omega^2 - \Omega^2(H_0)]}$$

(compare with[1]). Both measurements permit independent determinations of $(a_1 - b_1)^2$.

6. Let us examine a few more examples, with specific crystal structure and anisotropy taken into account. We limit outselves to the first power in the expansion of the coefficients of the equations of motion in the spin densities and consider a uniform deviation from equilibrium.

Take a vectorial antiferromagnet with crystallographic symmetry group D_{3d} (e.g., Cr_2O_3, Ti_2O_3). In the thermodynamic potential, among the terms of the fourth degree in the projections of the spin densities, we take only the large exchange terms and take no account of the hexagonal anisotropy:

$$\Phi = {}^1/_2 A\mathbf{L}^2 + {}^1/_2 B\mathbf{M}^2 + {}^1/_2\alpha L_z^2 + {}^1/_2\beta M_z^2 + {}^1/_2 C(\mathbf{LM})^2 + {}^1/_4 D_1\mathbf{L}^4 + {}^1/_2 D_2\mathbf{L}^2\mathbf{M}^2 - \mathbf{MH}. \tag{13}$$

We consider the state $\mathbf{H}_0 \parallel \mathbf{n}$, $\mathbf{L}_0 \perp \mathbf{n}$, which is stable only for $\alpha > 0$ or for $H_0 > H_1 \approx (|\alpha|\,BL_0)^{1/2}$. If we number $m_x, m_y, m_z, l_x, l_y, l_z$ by the numbers 1, 2, 3, 4, 5, 6, we can write

$$\begin{aligned} &\alpha_{11} = \eta + CL_{0x}^2, \quad \alpha_{12} = CL_x^0 L_y^0, \quad \alpha_{16} = CM_z^0 L_x^0;\\ &\alpha_{22} = \eta + CL_{0y}^2, \quad \alpha_{26} = CM_z^0 L_y^0, \quad \alpha_{33} = \eta + \beta,\\ &\alpha_{34} = 2D_2 L_x^0 M_z^0, \quad \alpha_{35} = 2D_2 L_y^0 M_z^0, \quad \alpha_{44} = 2D_1 L_x^{02},\\ &\alpha_{45} = 2D_1 L_x^0 L_y^0, \quad \alpha_{55} = 2D_1 L_{y0}^2, \quad \alpha_{66} = \alpha + CM_z^{02}, \end{aligned}$$

the remaining α_{ik} are zero ($\eta \approx B + D_2 L_0^2$).

As was expected, this leads to the antiferromagnet described by (13) having in all two resonance frequencies ($\Delta_1 = 0$). The Landau-Lifshitz equations yield only

one. However, if we use the thermodynamically allowable equations of motion the coefficients of which are written out to the first powers of the projections of $\mathbf{M}_0$ and $\mathbf{L}_0$ (cf. (6)):

$$\gamma_{12}=\rho_3 M_x^0,\quad \gamma_{14}=\rho_4 M_x^0,\quad \lambda_{14}=-\lambda_{25}=\lambda_1 L_x^0,\quad \lambda_{15}=-\lambda_1 L_y^0+\lambda_4 L_z^0,$$
$$\lambda_{16}=\lambda_7 L_y^0,$$
$$\gamma_{23}=\rho_4 M_x^0,\quad \lambda_{24}=-(\lambda_1 L_y^0+\lambda_4 L_z^0),$$
$$\lambda_{26}=-\lambda_7 L_x^0,\quad \lambda_{34}=\lambda_9 L_y^0,\quad \lambda_{35}=-\lambda_9 L_x^0,\quad \lambda_{36}=0,$$
$$\rho_{45}=\rho_1 M_z^0,\quad \rho_{46}=\rho_2 M_y^0,\quad \rho_{56}=-\rho_2 M_x^0, \tag{14}$$

then

$$\mathrm{Sp}\,K_1=\alpha\eta\lambda_7^2L_0^2+\eta^2\rho_3^2M_0^2+\eta CL_0^2(\lambda_7+\rho_3)^2M_0^2 + 2D_1\lambda_1^2[\eta L_0^4+C(L_{0x}^3-3L_{0x}L_{0y}^2)^2],$$
$$\mathrm{Sp}\,K_2=2D_1\eta\lambda_1^2\lambda_7^2[\alpha(\eta+CL_0^2)+\eta CM_0^2](L_{0x}^3-3L_{0x}L_{0y}^2)^2. \tag{15}$$

Comparing (14) with (15), we see that λ_1 is related to anisotropic interactions. From this it follows that the lower of the two resonance frequencies may have a gap with hexagonal anisotropy, even if the terms of the thermodynamic potential that determine the static hexagonal anisotropy are for any reason anomalously small.

To estimate the magnitude of this gap we write the answer for $H_0 = 0$:

$$\omega_{1,2}^2=\tfrac{1}{2}\{\alpha\eta\lambda_7^2L_0^2+2D_1\lambda_1^2(\eta+CL_0^2\cos^2 3\varphi_L)L_0^4 \pm([\alpha\eta\lambda_7^2L_0^2-2D_1\lambda_1^2(\eta+CL_0^2\cos^2 3\varphi_L)L_0^4]^2 + 8D_1\eta\lambda_1^2\lambda_7^2L_0^6\sin^2 3\varphi_L)^{1/2}\}.$$

Here $L_X^0 = L_0\cos\varphi_L$. The ratio

$$\omega_{1max}^2/\omega_{2min}^2\approx\lambda_1^2D_1L_0^2/\lambda_7^2\alpha\quad(\omega_2\sim 10^{13}sec)$$

is of order v^2/c^2. However, for sufficiently low temperatures, where the spin-wave approximation operates, we have $\lambda_1 \equiv 0$.

It is interesting to note that the lower frequency is the "third" in the sense that in none of the variables do the equations (6) with coefficients λ_{ik}, ρ_{ik}, and γ_{ik} of (14) decompose into two systems of equations like (11) and (12). In fact, in this case

$$\Delta_1=\lambda_1^2\lambda_7^2\lambda_9^2L_0^6\cos^2 3\varphi_L.$$

And if we go to decomposable systems, i.e., if we take the equations

$$\dot{s}=(b_1-a_1')\cos\xi_0 f_\varphi,\quad S_0\dot{\xi}=-(b_1+a_1'\,\mathrm{ctg}^2\,\xi_0)\sin\xi_0 f_\varphi,$$
$$\dot{\varphi}=-(b_1-a_1')\cos\xi_0 f_s+S_0^{-1}(b_1+a_1'\,\mathrm{ctg}^2\,\xi_0)\sin\xi_0 f_\xi,$$
$$S_0\cos\xi_0\dot{\chi}=(b_1-a_1)f_\gamma,\quad \cos\xi_0\dot{\theta}=b_1 f_\gamma,$$
$$S_0\cos\xi_0\dot{\gamma}=-(b_1-a_1)f_\chi-S_0b_1f_\theta,$$

then even with account taken of anisotropy in the coefficients we will have $\mathrm{Sp}\,K_2 = 0$, and the system has only one frequency of uniform resonance.

The fact that in weak ferromagnets the Goldstone branch of the spectrum is the "third," whereas in vectorial antiferromagnets it is the lower of the "old" resonance frequencies, is not accidental, but due to the symmetry of the ordering parameter[9]. Actually, the appearance in a weak ferromagnet of L_y^0 isolates in fact the direction $x(M_X^0)$, whereas in a vectorial antiferromagnet the y axis is isolated. Hence in the first case in the normal coordinates responsible for the Goldstone branch of the spectrum should appear $\Delta\chi$ and $\Delta(S\sin\xi)$, which belong to different "old" spectral branches, and in the second case, $\Delta\varphi$ and $\Delta(S\cos\xi)$, which belong to one "old" branch. But if we take the conditions $S^2 = \mathrm{const}$ and $\chi = \mathrm{const}$, the "perpendicular"[9] coordinate both in the first and in the second case becomes $\Delta\varphi$. If in the vectorial antiferromagnet $\mathbf{H}_0 \perp \mathbf{n}$, then, as in the weak ferromagnet, a bicubic equation must be solved to investigate the third eigenfrequency. However, all that has been said about anisotropy of the lower spectral branch remains in force, since in this case ($M_X = M_0\cos\varphi_M$),

$$\Delta_2=\lambda_1^2\{M_0^2L_0\rho_2\rho_4\cos(2\varphi_M+\varphi_L)+L_0^3\lambda_7\lambda_9\cos 3\varphi_L\}^2.$$

7. The dependence of the "third" frequency on H_0 is easily calculated for the vectorial antiferromagnet when $\mathbf{L} \parallel \mathbf{H}_0 \parallel \mathbf{n}$. Let $G = C_{ni}$ and the magnetic symmetry group be $C_n \times IT$. The addition to Φ on account of deviation from the equilibrium configuration is determined by the following nonzero α_{ik}:

$$\alpha_{11}=\alpha_{22}=\tfrac{1}{2}\varkappa_{\perp 0}^{-1},\quad \alpha_{33}=\tfrac{1}{2}\varkappa_{\parallel 0}^{-1},\quad \alpha_{44}=\alpha_{55}=\tfrac{1}{2}(\alpha+C\varkappa_{\parallel 0}^{-1}H_0^2),$$
$$\alpha_{66}=2D_1L_0^2,\quad \alpha_{14}=\alpha_{25}=\varkappa_{\perp 0}^{-1}CL_0H_0,\quad \alpha_{36}=\varkappa_{\parallel 0}^{-1}(C+D_2)L_0H_0,$$

where $\varkappa_{\parallel 0}^{-1}=B+\beta+(C+D_2)L_0^2$, $\varkappa_{\perp 0}^{-1}=B+D_2L_0^2$.

The thermodynamically allowable equations of motion split into two groups in accordance with the uniaxiality of the crystal. For the perpendicular projections $\gamma_{12} = \gamma_1 M_0$, $\rho_{45} = \rho_1 M_0$, $\lambda_{14} = \lambda_{25} = \lambda_3 L_0$, $\lambda_{15} = -\lambda_{24} = \lambda_4 L_0$ and for longitudinal $\lambda_{36} = \lambda_5 L_0$. Comparing with (5) we see that λ_5 is of relativistic origin. The frequency of uniform oscillations associated with longitudinal projections l and m is excited by parallel pumping and equals

$$\omega_3^2=2\lambda_5^2L_0^2\{\varkappa_\parallel^{-1}D_1L_0^2-2(C+D_2)^2L_0^2\varkappa_{\parallel 0}^{-2}H_0^2\},$$

i.e., it decreases quadratically with increasing dc field. (This configuration is stable to fields $H_0 \le (|\alpha|/CL_0^2)^{1/2}\varkappa_{\parallel 0}^{1/2}L_0$.) The ratio of ω_3 to the frequency of transverse oscillations usually considered in spin wave theory at $H_0 = 0$ is

$$\frac{\omega_3^2}{\omega_{1,2}^2}\approx\frac{2\lambda_5^2D_1L_0^2}{(\lambda_3+\lambda_4)^2\alpha}\frac{\varkappa_\perp^0}{\varkappa_\parallel^0}\sim\frac{v^2}{c^2}.$$

However, in seeking the third branch of the spectrum in this case it is important to remember that the decay of the branch is due to large exchange interactions. In this configuration we see that the new frequency is completely analogous to second sound waves passing through the sublattices in opposite directions—a second sound standing wave[14,15]. The phenomenological parameter Φ_{11} that figures everywhere in the Goldstone branch in the case of a pure antiferromagnet (when a solution of Type I according to[8] is realized) is easily related to the heat capacity of the magnetic subsystem: $c_H = -t(\partial S^2/\partial t)^2\Phi_{11}$ (t is the temperature). In other situations the heat capacity of the magnetic subsystem is associated with combinations of second derivatives.

Everywhere in the final estimates in Secs. 6 and 7 it was assumed that the coefficients of the equations of motion having relativistic origin are quadratic in the spin-orbit interaction, as are the relativistic constants in Φ. However, this assumption requires justification and a microscopic model for calculation of λ_1 and λ_5 is needed. This is outside the scope of the present paper. Relativistic corrections to the exchange g fac-

tor of one sublattice are of first order in spin-orbit interactions[16].

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