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The phenomenological theory of magnetic resonance and of spin waves in antiferromagnetics

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We construct a theory of magnetic resonance in antiferromagnets. The theory contains the maximum possible number of independent phenomenological functions of the temperature that can be determined in principle from dynamic and static experiments. In the high-temperature region the equations of the theory are essentially those of Onsager's theory of thermodynamic fluctuations. At low temperatures, in the so-called spin wave region, several parameters of the Onsager theory turn out to be the same. We describe this situation by new equations for magnetic resonance which are second-order equations in time and which can be derived by means of Lagrange's mechanical principle. As actual cases we consider the most lucid cases of a two-sublattice antiferromagnet in an external field and of the antiferromagnets $CoCO_3$, $MnCO_3$, $FeCO_3$, CoF_2, and MnF_2.

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1. INTRODUCTION

When studying magnetic resonance and spin waves in antiferromagnets theoretically or experimentally it is extremely useful to have general phenomenological expressions for the spectra which contain the maximum number of constants compatible with the symmetry and physical properties of the phenomenon. The first step along this path was the famous Landau–Lifshitz equation[1] which describes ferromagnetic resonance and the essentially long-wavelength spin waves in a ferromagnet. The first phenomenological theory of antiferromagnetic resonance (AFMR) was the theory of Kittel and Keffer[2] (see also[3]) constructed in the molecular field approximation. Borovik-Romanov and Turov (see[4,5] and also[6]) developed a phenomenological theory of AFMR in the large spin approximation for $T=0$. The first attempt to construct a theory of AFMR containing the maximum possible number of constants was undertaken by Gufan.[7]

An essential step in the range of phenomenological theories was the hydrodynamic theory of spin waves constructed by Halperin, Hohenberg, and others.[8] Finally, a complete phenomenological theory of spin waves in the exchange approximation and for the case where there are no external fields was very recently produced by Andreev and Marchenko.[9] Essentially they determined all possible kinds of "acoustic" oscillations in a magnetic system. It turned out that while for oscillations of atoms there are always three and only three acoustic branches, for oscillations of magnetic moments the number of acoustic modes can be both less and more than three. It is determined by the specific exchange group of magnetic symmetry introduced by the authors. Exchange magnetic groups differ from the magnetic groups (including time-reversal) introduced by Landau[10] (see also[11]) in his time and are the same as the so-called color groups.

In the high-temperature range $T \lesssim T_c$ (T_c is the magnetic phase-transition temperature) the theory of AFMR can be constructed in a natural way as a particular case of the general Onsager theory of thermodynamic fluctuations (see, e.g.,[12]). In fact, one can write the AFMR equations as Onsager equations for thermodynamic variables:

$$\frac{dq_a}{dt} = -\sum_b \gamma_{ab} \frac{\partial f}{\partial q_b}. \qquad (1)$$

Here f is the free energy of a small deviation from equilibrium which is, in general, an arbitrary positive definite quadratic form of the variables q_a. The coefficients γ_{ab} are the Onsager coefficients; their form is restricted only by the symmetry principle of the kinetic coefficients. It is clear that if the motion described by Eq. (1) is undamped, f must be an integral of the motion and the coefficients γ_{ab} must therefore be antisymmetric: $\gamma_{ab} = -\gamma_{ba}$. The symmetric part of γ_{ab} describes damping.

The thermodynamic variables q_a themselves can be introduced in two ways. We can neglect in first approximation the lattice deformations connected with magnetostriction and we can consider the motion of the system of spins against the background of the spatial symmetry of the paramagnetic phase (this method has been used earlier by Gufan and one of the present authors[13]). In such an approach the set of quantities q_a will be the same as the set of "vectors" $\mathbf{S}_{n\alpha}$, where the vector index i of $S^i_{n\alpha}$ describes the direction of the spins in space while the set of indexes $n\alpha$ describes the coordinate dependence of the spin density. In our case n is the number of the irreducible representation of the space group of the paramagnetic phase, while α is the number of the function in its base. On the other hand, we can understand by the irreducible representation n either a representation of Landau's magnetic group in the magnetic phase[10] or a representation of the exchange group introduced by Andreev and Marchenko.[9] In the first case, however, it is difficult in practice to separate relativistic from exchange effects while in the second case there remains the unsolved problem of how to find the exchange group itself from the available experimental data.[1)]

The AFMR frequencies given by Eqs. (1) are clearly determined both by the static properties of the system such as the spin-flip fields H_0 and the perpendicular $\chi_\perp$ and parallel $\chi_\parallel$ susceptibilities which occur in f, and by the dynamic coefficients γ_{ab}. However, a comparison with the results of a microscopic theory (e.g., with Oguchi's theory[14]) and with a phenomenological spin wave theory (see Sec. 4 below) shows that at low temperatures in the spin wave region not all coefficients γ_{ab} allowed by Onsager's principle are independent. The fact is that, as was first shown by Dyson,[15] there are two kinds of temperature dependence for all physical quantities for $T < T_c$: a series expansion in powers of T/T_c and a dependence of the form $\exp(-T_c/T)$. The region $T \ll T_c$ where only the power-law temperature dependence remains while all $\exp(-T_c/T)$ are negligibly small is also the spin wave region. Knowing the behavior of a quantity only in the spin wave region we can therefore say nothing whatever about the region $T \sim T_c$. However, in return we can conclude from the results of the spin wave theory which of the quantities γ_{ab} or of their combinations turn to zero as $\exp(-T_c/T)$.

Of course, we can always turn directly to the microscopic theory. However, concrete microscopic calculations taking into account anisotropy and external differently orientated fields, e.g., in the framework of Holstein and Primskoff's method (see, e.g.,[14]) are extremely laborious. It is therefore always useful to have a phenomenological method in which the part of the microscopic theory is reduced to merely calculating the temperature dependence of two or three coefficients. Moreover (see the end of Sec. 4) a phenomenological method enables us to make a number of generalizations which go beyond the framework of present-day microscopic theories which operate in the limit of large spins S.

The phenomenological method proposed by us is essentially a generalization of the usual Lagrangian method in the small oscillations theory. We shall consider the free energy f of the oscillations, which occurs in (1), as a potential energy and we shall introduce apart from it a Lagrangian function $\mathscr{L}$. In contrast to the standard situation in the theory of oscillations there can now occur in $\mathscr{L}$, in the case when the variables are magnetic moments, i.e., quantities which change sign under time reversal, also terms which are linear in the derivatives.

The general expression for $\mathscr{L}$ is of the form

$$\mathscr{L} = \frac{1}{2}\sum_{ab} \rho_{ab} q_a \frac{dq_b}{dt} + \frac{1}{2}\sum_{ab} \mu_{ab} \frac{dq_a}{dt}\frac{dq_b}{dt} - f. \tag{2}$$

The coefficients ρ_{ab} must be antisymmetric: $\rho_{ab} = -\rho_{ba}$, in order that the first term in (2) does not reduce to a total derivative with respect to the time. Since, as is clear from (2), the ρ_{ab} must change sign as $t \to -t$, they are, like the γ_{ab} in (1), proportional to odd powers of the equilibrium values of the spin density (see Secs. 3, 4).

The equations of motion are now the Lagrangian equations ($\dot{q} \equiv dq/dt$) which with the $\mathscr{L}$ from (2) have the form

$$\sum_b \rho_{ba} \dot{q}_b + \sum_b \mu_{ab} \ddot{q}_b = -\frac{\partial f}{\partial q_a}. \tag{3}$$

At first sight this equation (at least when $\mu = 0$) is Eq. (1) solved for $\partial f/\partial q_a$. However, in actual fact the tensor ρ_{ab} (and γ_{ab}) may not have an inverse and, as always, Eq. (3) is equivalent to a set of first-order equations with double the number of variables (coordinates and momenta), i.e., Hamiltonian equations, and not Eqs. (1).

Equation (3) has an energy integral $\mathscr{E}$, in our case the spin wave energy

$$\mathscr{E} = \sum_a \dot{q}_a \frac{\partial \mathscr{L}}{\partial \dot{q}_a} - \mathscr{L} = \frac{1}{2}\sum_{ab} \mu_{ab} \dot{q}_a \dot{q}_b + f. \tag{4}$$

which equals the sum of kinetic and potential energies. Equation (4) requires that the tensor μ_{ab} be positive definite.

Apart from the energy of the excitation $\mathscr{E}$ we can also determine its (spin) angular momentum $\mathbf{J}$. To do this, as always in mechanics (see, e.g.,[16] Sec. 43), we must consider the action

$$\mathscr{S} = \int^t \mathscr{L}\, dt$$

as function of the coordinates and recognize that when we vary the endpoint of the trajectory $\delta q_a(t)$ the action changes according to the formula

$$\delta\mathcal{S}=\sum_a \frac{\partial\mathcal{L}}{\partial\dot q_a}\delta q_a. \tag{5}$$

We consider now how the action changes when all spins i.e., the above introduced quantities $\mathbf{S}_{n\alpha}$, are rotated over the same infinitesimal angle $\delta\varphi$. Equation (5) becomes in that case

$$\delta\mathcal{S}=\mathbf{J}\delta\boldsymbol{\varphi}, \tag{6}$$

where, by definition, **J** is also the angular momentum.

In the case when the angular momentum is an integral of motion, i.e., when the Lagrangian function $\mathcal{L}$ does not change under rotation around any axis (this means an exchange interaction or that the crystal is magnetically uniaxial) it can be connected with the motion of the magnetic moment $\mathcal{M}$, averaged over a period, by introducing the magnetomechanical ratio γ_* for the excitation:

$$\bar{\mathcal{M}}=\gamma_* J. \tag{7}$$

On the other hand, we can evaluate the magnetic moment $\mathcal{M}(t)$ as the derivative with respect to the external magnetic field H using the normal mechanics formula (see[16], Sec. 40)

$$\mathcal{M}(t)=\left(\frac{\partial\mathcal{L}}{\partial H}\right)_{q,\dot q}=-\left(\frac{\partial\mathcal{H}}{\partial H}\right)_{q,p}, \tag{8}$$

where the derivatives of the Lagrangian function $\mathcal{L}$ are taken for constant coordinates q and velocities $\dot q$, while the derivatives of the Hamiltonian function $\mathcal{H}$ are for constant q and their conjugate momenta p. We shall see below in Sec. 4 that Eq. (7) imposes well defined limitations on the coefficients ρ and μ in (2).

The damping of the AFMR is in the Onsager version (1) of the theory evaluated, as always, by using the positive definite entropy generation:

$$T\frac{ds}{dt}=-\frac{1}{2}\sum_{ab}\gamma_{ab}{}^{s}q_a q_b,$$

where γ^s_{ab} is the symmetric part of the Onsager coefficients. For small damping we can always use the relation

$$\frac{df}{dt}=-T\frac{ds}{dt}, \quad \text{or} \quad \frac{2}{\tau}=T\frac{d\bar s}{dt}\Big/ f,$$

which is averaged over a period and which also determines the relaxation time τ.

In the spin wave version a positive dissipation function $\mathcal{R}$ (see[16], Sec. 25) is introduced

$$\mathcal{R}=\frac{1}{2}\sum_{ab}\lambda_{ab}\dot q_a\dot q_b,$$

$$\frac{d}{dt}\frac{\partial\mathcal{L}}{\partial\dot q_a}-\frac{\partial\mathcal{L}}{\partial q_a}=-\frac{\partial\mathcal{R}}{\partial\dot q_a} \tag{9}$$

while the relaxation time τ is determined from the relations

$$\frac{d\mathcal{E}}{dt}=-\mathcal{R} \quad \text{or} \quad \frac{2}{\tau}=\frac{\overline{\mathcal{R}}}{\mathcal{E}}. \tag{10}$$

We apply in what follows the proposed general scheme to the simplest case of a two-sublattice uniaxial antiferromagnetic in an external field. Of course, the scheme with a Lagrangian function is applicable to spin waves with a finite wavevector; to do this we must in the usual manner take into account terms with derivatives with respect to the coordinates in f in (2) and in $\mathcal{R}$ in (9). Equations, similar to (9) and (1) then completely determine the momentum dependence of the spin wave damping.

2. THERMODYNAMIC RELATIONS

We start with a calculation of the free energy f of the oscillations which occurs in both versions of the theory, both the high-temperature and the spin-wave one. We consider a uniaxial ferromagnetic and a uniaxial two-sublattice antiferromagnetic in an external magnetic field H. We then restrict ourselves to the first approximation in the anisotropy a and in H and we shall neglect the anisotropy in the basis plane.

In the ferromagnetic case the free energy is of the form (z is here and henceforth the crystal axis)

$$F=F_0(M^2)-{}^1\!/_2 a(M^2)M_z^2-\mathbf{MH}, \tag{11}$$

where **M** is the magnetic moment per unit volume, $F_0(M^2)$ the exchange part of the free energy, $a(M^2)$ the anisotropy energy which, in general, is a function of M^2. The value of the moment **M** in equilibrium can be found from the condition

$$\frac{\partial F}{\partial\mathbf{M}}=0. \tag{12}$$

One can also find easily the energy f connected with a small deviation **m** of the moment from equilibrium. In the "easy axis" case and the field H along z we have$^{2)}$

$$f={}^1\!/_2(a+H/M)\mathbf{m}_\perp^2+{}^1\!/_2\chi_\parallel^{-1}m_z^2. \tag{13}$$

Here a and M are given functions of the temperature, $\chi_\parallel^{-1}$ is the susceptibility of the paraprocess, and

$$\chi_\perp^{-1}=a+H/M \tag{14}$$

is the perpendicular susceptibility of the uniaxial ferromagnet. It depends, of course, on the field and on a.

The state of the simplest two-sublattice antiferromagnet in an external magnetic field is determined by two vectors $\mathbf{S}_{n\alpha}$ mentioned in the Introduction, viz., the antiferromagnetic vector **L** and the magnetic moment **M**. For a uniaxial crystal when there is no anisotropy in the basis plane we can write for the free energy

$$F=F_0(L^2)+{}^1\!/_2P(L^2)(\mathbf{LM})^2+{}^1\!/_2Q(L^2)\mathbf{M}^2-{}^1\!/_2a(L^2)L_z^2-\mathbf{MH}. \tag{15}$$

We retained in Eq. (15) solely the quadratic terms in the expansion of the free energy in terms of the quantity M which is small for an antiferromagnet. This corresponds to the main approximation in powers of the ratio H/H_e of the external field H to the exchange field H_e. Moreover, we neglected the intrinsic relativistic anisotropy of the magnetic susceptibility (i.e., terms of the form M_z^2 which give the anisotropy of the susceptibility in the paramagnetic phase). As in the ferromagnetic case, all quantities F_0, P, Q, and a which occur in (15) are arbitrary functions of L^2.

The equilibrium values of **L** and **M** are determined from the conditions

$$\frac{\partial F}{\partial \mathbf{L}}=\frac{\partial F}{\partial \mathbf{M}}=0,$$

and the second of these gives, as always,

$$\mathbf{M}=\chi_\perp\mathbf{H}+(\chi_\parallel-\chi_\perp)\mathbf{L}(\mathbf{HL})/L^2,$$
$$\chi_\perp^{-1}=Q,\quad \chi_\parallel^{-1}-\chi_\perp^{-1}=PL^2, \tag{16}$$

where $\chi_\parallel$ and $\chi_\perp$ are the parallel and perpendicular susceptibilities defined in the usual way, neglecting relativistic effects.

The formulae for the free energy of the oscillations of **l** and **m** depend on the actual situation.

a) *Easy axis*, $\mathbf{H}\parallel\mathbf{z}$. We must here distinguish two cases: weak field $H<H_0$ (H_0 the spin-flip field, see below) and strong field.

In a weak field $H<H_0$ and the antiferromagnetic vector $\mathbf{L}\parallel\mathbf{z}$. In that case

$$f=\frac{1}{2}\left\{a-\frac{\chi_\parallel H^2}{L^2}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\right\}\mathbf{l}_\perp^2+\frac{H}{L}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\mathbf{l}_\perp\mathbf{m}_\perp$$
$$+\frac{1}{2}\chi_\perp^{-1}\mathbf{m}_\perp^2+\frac{1}{2}Al_z^2+BHl_zm_z+\frac{1}{2}\chi_\parallel^{-1}m_z^2. \tag{17}$$

The constants A and B which determine the energy of the longitudinal oscillations do not have a direct experimental meaning and can be expressed in a well defined way in terms of derivatives of F_0, P, Q, and so on.

The condition that the energy of the transverse oscillations be positive definite has the form

$$H^2\leqslant H_0^2=aL^2/(\chi_\perp-\chi_\parallel), \tag{18}$$

where H_0 is the above-mentioned thermodynamic spin-flip field. The condition for stability with respect to longitudinal perturbations $B^2H^2\leqslant A/\chi_\parallel$ is normally used together with (18).

In a strong field $H>H_0$ and $\mathbf{L}\parallel\mathbf{x}$. In that case

$$f=\frac{1}{2}\left\{-a+\frac{\chi_\perp^2H^2}{\chi_\parallel L^2}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\right\}l_z^2+\frac{H}{L}\frac{\chi_\perp}{\chi_\parallel}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)l_zm_x$$
$$+{}^1/_2\chi_\parallel^{-1}m_x^2+{}^1/_2\chi_\perp^{-1}m_y^2+{}^1/_2Al_x^2+BHl_xm_z+{}^1/_2\chi_\perp^{-1}m_z^2. \tag{19}$$

The energy f is positive in fields $H>H_0$, but such that $B^2H^2<A/\chi_\perp$. Usually the second condition is also satisfied.

b) *Easy axis*, $\mathbf{H}\parallel\mathbf{x}$. In that case

$$f=\frac{1}{2}\left\{a+\frac{\chi_\perp^2H^2}{\chi_\parallel L^2}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\right\}l_x^2+\frac{1}{2}al_y^2+\frac{H}{L}\frac{\chi_\perp}{\chi_\parallel}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)l_xm_z$$
$$+{}^1/_2\chi_\perp^{-1}m_y^2+{}^1/_2\chi_\parallel^{-1}m_z^2+{}^1/_2Al_z^2+BHl_zm_x+{}^1/_2\chi_\perp^{-1}m_x^2. \tag{20}$$

c) *Easy plane*, $\mathbf{L}\parallel\mathbf{x}$, $\mathbf{H}\parallel\mathbf{z}$. In that case Eq. (19) with $a<0$ is, of course, valid.

d) *Easy plane*, $\mathbf{L}\parallel\mathbf{x}$, $\mathbf{H}\parallel\mathbf{y}$. In that case

$$f=\frac{\chi_\perp^2H^2}{2\chi_\parallel L^2}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)l_y^2+{}^1/_2|a|l_z^2+\frac{H}{L}\frac{\chi_\perp}{\chi_\parallel}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)l_ym_x$$
$$+{}^1/_2\chi_\parallel^{-1}m_x^2+{}^1/_2\chi_\perp^{-1}m_z^2+{}^1/_2Al_x^2+BHl_xm_y+{}^1/_2\chi_\perp^{-1}m_y^2. \tag{21}$$

3. HIGH TEMPERATURES

We start with the ferromagnetic case. It is clear that in the case of a magnetic field which is parallel to the easy axis the longitudinal oscillations can be split from the transverse ones. As always the longitudinal ones appear to be purely relaxational and we shall not consider them. On the other hand, it follows from Eqs. (13) and (1) that the frequencies will be small, proportional to $H\sim a$. In the main approximation in a and H we must thus restrict ourselves to the exchange approximation for the coefficients γ_{ab}. This gives at once the simple equation

$$\frac{d\mathbf{m}_\perp}{dt}=\gamma\left[\mathbf{M}\times\frac{\partial f}{\partial \mathbf{m}_\perp}\right], \tag{22}$$

i.e., the linearized Landau–Lifshitz equation[1] and the well known expression for the ferromagnetic resonance frequency

$$\omega=\gamma(H+aM)=\gamma M\chi_\perp^{-1}. \tag{23}$$

The resonance frequency is thus determined by means of the quantities M, a, and $\chi_\perp^{-1}$ which are measured in static experiments and one new dynamic constant, the magnetomechanical ratio γ.

In the AFMR case comparison of Eqs. (17) to (21) with Eqs. (1) shows that the AFMR frequencies are again small and proportional to $\sim H$, $a^{1/2}$. We must here take into account that it can be seen from the formula for the energy f that in any mode the oscillating magnetic moment m is less than the corresponding antiferromagnetic vector l, or more precisely, $m\sim Hl$, $a^{1/2}l$. It is therefore also in this case sufficient to restrict oneself to the exchange approximation for the coefficients γ_{ab}.

The general equations with exchange values of γ_{ab} have the form

$$\frac{d\mathbf{l}}{dt}=\gamma_1\left[\mathbf{L}\times\frac{\partial f}{\partial \mathbf{m}}\right]+(\gamma_2-\gamma_4)(\mathbf{LM})\left[\mathbf{L}\times\frac{\partial f}{\partial \mathbf{l}}\right]+\gamma_4L^2\left[\mathbf{M}\times\frac{\partial f}{\partial \mathbf{l}}\right],$$
$$\frac{d\mathbf{m}}{dt}=\gamma_1\left[\mathbf{L}\times\frac{\partial f}{\partial \mathbf{l}}\right]+(\gamma_3-\gamma_5)(\mathbf{LM})\left[\mathbf{L}\times\frac{\partial f}{\partial \mathbf{m}}\right]+\gamma_5L^2\left[\mathbf{M}\times\frac{\partial f}{\partial \mathbf{m}}\right], \tag{24}$$

with, in general, five arbitrary "magnetomechanical ratios" γ.

To find the spectrum of Eq. (24) it is impossible to allow extra accuracy and we must drop all corrections of relative order H/H_e and H_a/H_e (H_a is the anisotropy field). In particular, the terms proportional to γ_2 and γ_4 in the first of Eqs. (24) are such corrections.

We now give the formulae for AFMR frequencies.

a) *Easy axis*, $\mathbf{H}\parallel\mathbf{z}$, $H<H_0$. There are two frequencies:

$$\omega_{1,2}=\pm\gamma_\parallel H+[H_0^2\gamma_*^2+H^2(\gamma_\parallel^2-\gamma_*^2)]^{1/2}. \tag{25}$$

Here H_0 is the thermodynamic spin-flip field (18) and we have introduced instead of the coefficients γ_1 and γ_3 new γ_* and $\gamma_\parallel$ through the formulae

$$\gamma_*^2=\gamma_1^2\left(1-\frac{\chi_\parallel}{\chi_\perp}\right), \quad \gamma_\parallel=\gamma_1\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)+\frac{1}{2}\gamma_3L^2\frac{\chi_\parallel}{\chi_\perp}. \tag{26}$$

In the molecular field model we have the relation

$$\gamma_1=\gamma_3L^2=\gamma_5L^2=\gamma_0, \tag{27}$$

where $\gamma_0=g\mu_0$ is the magnetogyric ratio for a free spin, and (25) becomes Kittel and Keffer's well known formula[2, 3]

$$\frac{\omega_{1,2}}{\gamma_0}=\pm H\left(1-\frac{\chi_\parallel}{2\chi_\perp}\right)+\left[\frac{aL^2}{\chi_\perp}+H^2\frac{\chi_\parallel^2}{4\chi_\perp^2}\right]^{1/2}. \tag{28}$$

In a strong field $H>H_0$, $\mathbf{L}\parallel\mathbf{x}$ and

$$\omega_1=[-H_0^2\gamma_*^2+\gamma_\perp^2H^2]^{1/2}, \quad \omega_2=0, \tag{29}$$

where instead of the constant γ_5 we have introduced a new magnetogyric ratio $\gamma_\perp$:

$$\gamma_\perp^2=\gamma_1^2\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)+\frac{\chi_\perp}{\chi_\parallel}\left(\gamma_5L^2-\gamma_1\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\right)^2. \tag{30}$$

In the molecular field limit $\gamma_\perp=\gamma_0$.

b) *Easy axis*, $\mathbf{H}\parallel\mathbf{x}$.

$$\omega_1=[H_0^2\gamma_*^2+\gamma_\perp^2H^2]^{1/2}, \quad \omega_2=H_0\gamma_*. \tag{31}$$

With the above indicated accuracy the frequency of the second branch is independent of the magnetic field.

c) *Easy plane*, $\mathbf{H}\parallel\mathbf{z}$. Equations (29) remain valid for this case, but we must substitute for the term $-H_0^2\gamma_*^2$: $|a|L^2\gamma_*^2/(\chi_\perp-\chi_\parallel)\equiv\gamma_1^2|a|L^2/\chi_\perp$.

d) *Easy plane*, $\mathbf{H}\parallel\mathbf{y}$. We have

$$\omega_1=\gamma_\perp H, \quad \omega_2=\frac{|a|L^2\gamma_*^2}{\chi_\perp-\chi_\parallel}=\frac{\gamma_1^2|a|L^2}{\chi_\perp}. \tag{32}$$

The formulae for the AFMR frequencies are thus in the high-temperature region of the same structure as the formulae from the molecular field theory (see, e.g., [3]). However, there now occurs in them instead of a single dynamical constant γ_0 three phenomenological constants: γ_*, $\gamma_\parallel$, and $\gamma_\perp$.

4. LOW TEMPERATURES

As in the high-temperature case the ferromagnetic resonance (FMR) and antiferromagnetic resonance (AFMR) frequencies will be small: $\omega\sim a$, H for FMR and $\omega\sim a^{1/2}$, H for AFMR. Moreover, the estimates for the amplitudes of the oscillations, $m\sim Hl$, $a^{1/2}l$ also remain valid. It is therefore again sufficient to write the kinetic part of the Lagrangian function $\mathscr{L}$ in the exchange approximation.

In the ferromagnetic case

$$\mathscr{L}=\frac{1}{2}\rho\mathbf{M}\left[\mathbf{m}\times\frac{d\mathbf{m}}{dt}\right]+\frac{1}{2}(\mu-\mu_1)\left(\mathbf{M}\frac{d\mathbf{m}}{dt}\right)^2$$
$$+\frac{1}{2}\mu_1M^2\left(\frac{d\mathbf{m}}{dt}\right)^2-\frac{1}{2}\left(a+\frac{H}{M}\right)m_\perp^2-\frac{1}{2}\chi_\parallel^{-1}m_z^2. \tag{33}$$

In the equation of motion for the transverse components the term with the second derivative is a small correction and the low-temperature equations give the same result as the Landau-Lifshitz Eq. (22) with

$$\gamma=-1/\rho M^2. \tag{34}$$

In the equation of motion for the longitudinal mode the term with the second derivative must be retained:

$$\mu M^2\frac{d^2m_z}{dt^2}+\frac{1}{\chi_\parallel}m_z=-\lambda\frac{dm_z}{dt}. \tag{35}$$

In the right-hand side of this equation we included the general form of the relaxation term given by the dissipation function $\mathscr{R}$. However, the value of this equation is not particularly large as in actual fact it only describes a relaxation mode. This is not ultimately connected with the large magnitude of $\chi_\parallel$ as already in the Bloch model of spin waves $\chi_\parallel\sim T/H^{1/2}$, $T/a^{1/2}$ (for details see, e.g., [17]).

In the antiferromagnetic case the Lagrangian function has the form

$$\mathscr{L}=\rho\mathbf{L}\left[\mathbf{l}\times\frac{d\mathbf{m}}{dt}\right]+(\rho_1-\rho_2)(\mathbf{LM})\mathbf{L}\left[\mathbf{l}\times\frac{d\mathbf{l}}{dt}\right]$$
$$+\rho_2L^2\mathbf{M}\left[\mathbf{l}\times\frac{d\mathbf{l}}{dt}\right]+\frac{1}{2}\mu L^2\left(\frac{d\mathbf{l}}{dt}\right)^2+\frac{1}{2}(\mu_1-\mu)\left(\mathbf{L}\frac{d\mathbf{l}}{dt}\right)^2-f, \tag{36}$$

where f is in the various cases given by the formulae from Sec. 2. When writing down Eq. (36) we took into account the above mentioned relations between the oscillation amplitudes $\mathbf{m}$ and $\mathbf{l}$ and dropped small exchange terms of the kind

$$\mathbf{M}\left[\mathbf{m}\times\frac{d\mathbf{m}}{dt}\right], \quad \left(\frac{d\mathbf{m}}{dt}\right)^2, \quad \left(\mathbf{L}\frac{d\mathbf{m}}{dt}\right)\left(\mathbf{M}\frac{d\mathbf{l}}{dt}\right)$$

and so on.

We now find the ratios γ_*, mentioned in the Introduction, between the coefficients in the kinetic part of the Lagrangian $\mathscr{L}$ and the magnetomechanical moment for the excitation from Eq. (7). We start with the simplest ferromagnetic case. The Lagrangian $\mathscr{L}$ of the system is invariant under a rotation of all transverse moments $\mathbf{m}_\perp$ (in what follows we denote $\mathbf{m}_\perp$ simply by $\mathbf{m}$) over a small angle $\delta\varphi$ around the z-axis:

$$\delta\mathbf{m}=[\mathbf{z}\times\mathbf{m}]\,\delta\varphi.$$

In accordance with Eqs. (5), (6) to this rotation there corresponds a conserved angular momentum of the oscillation (of course directed along the z-axis):

$$J={}^1\!/_2\rho M\mathbf{m}^2. \tag{37}$$

On the other hand, evaluating the magnetic moment $\mathscr{M}$ using Eq. (8) gives

$$\mathscr{M}=-\mathbf{m}^2/2M. \tag{38}$$

As the magnetic moment $\mathscr{M}$ itself is according to (38), (37) a conserved quantity, direct comparison with Eq. (7) gives

$$\gamma_*=-1/\rho M^2.$$

Comparison with (34) shows that the constant γ in the Landau–Lifshitz equation is the same as the magnetomechanical ratio γ_* for a spin wave.

We now consider an antiferromagnetic with an easy axis in a weak field ($H<H_0$), parallel to the z-axis. The Lagrangian function for transverse oscillations[3)] has the form

$$\mathscr{L}=\rho L\left[\mathbf{l}\times\frac{d\mathbf{m}}{dt}\right]\mathbf{z}+\rho_1L^2\chi_\parallel H\left[\mathbf{l}\times\frac{d\mathbf{l}}{dt}\right]\mathbf{z}+\frac{1}{2}\mu L^2\left(\frac{d\mathbf{l}}{dt}\right)^2$$
$$-\frac{1}{2}\left\{a-\frac{\chi_\parallel H^2}{L^2}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\right\}\mathbf{l}^2-\frac{H}{L}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\mathbf{lm}-\frac{1}{2}\chi_\perp{}^{-1}\mathbf{m}^2. \tag{39}$$

It is invariant under a rotation of $\mathbf{l}$ and $\mathbf{m}$ around the z-axis over the same angle $\delta\varphi$:

$$\delta\mathbf{m}=[\mathbf{z}\times\mathbf{m}]\delta\varphi,\quad \delta\mathbf{l}=[\mathbf{z}\times\mathbf{l}]\delta\varphi,$$

which according to (5), (6) gives the following formula for the conserved angular momentum of the oscillation:

$$J=\rho L\mathbf{ml}+\rho_1L^2\chi_\parallel H\mathbf{l}^2+\mu L^2\left[\mathbf{l}\times\frac{d\mathbf{l}}{dt}\right]\mathbf{z}. \tag{40}$$

On the other hand, the use of Eq. (8) to evaluate the magnetic moment leads to

$$\mathscr{M}(t)=\rho_1L^2\chi_\parallel\left[\mathbf{l}\times\frac{d\mathbf{l}}{dt}\right]\mathbf{z}+\frac{\chi_\parallel H}{L^2}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\mathbf{l}^2-\frac{1}{L}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\mathbf{lm}. \tag{41}$$

In order to apply now Eq. (8) we must average (41) over a period of the oscillations. To do this we write down the equations of motion defined by (39):

$$\rho L\left[\mathbf{z}\times\frac{d\mathbf{l}}{dt}\right]=-\frac{1}{\chi_\perp}\mathbf{m}-\frac{H}{L}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\mathbf{l},$$
$$\rho L\left[\mathbf{z}\times\frac{d\mathbf{m}}{dt}\right]+2\rho_1L^2\chi_\parallel H\left[\mathbf{z}\times\frac{d\mathbf{l}}{dt}\right]+\mu L^2\frac{d^2\mathbf{l}}{dt^2}$$
$$=-\left\{a-\frac{\chi_\parallel H^2}{L^2}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\right\}\mathbf{l}-\frac{H}{L}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right)\mathbf{m}. \tag{42}$$

Using them we can reduce Eqs. (40) and (41) for J and $\mathscr{M}(t)$ to the form

$$J=(\chi_\perp\rho^2+\mu)L^2\left[\mathbf{l}\times\frac{d\mathbf{l}}{dt}\right]\mathbf{z}+H\{-\rho(\chi_\perp-\chi_\parallel)+\rho_1L^2\chi_\parallel\}\mathbf{l}^2,$$

$$\mathscr{M}(t)=\{-\rho(\chi_\perp-\gamma_\parallel)+\rho_1L^2\chi_\parallel\}\left[\mathbf{l}\times\frac{d\mathbf{l}}{dt}\right]\mathbf{z}+\frac{H}{L^2}(\chi_\perp-\chi_\parallel)\mathbf{l}^2. \tag{43}$$

A comparison of $\mathscr{M}$ with γ_*J in (43) gives in fact two relations as $\mathscr{M}$ and J are functions of the magnetic field H. In particular, the fact that $\mathscr{M}$ is the same as γ_*J for $H=0$ leads automatically to the fact that the coefficients of $\mathbf{I}^2$ and of $[\mathbf{Il}]\mathbf{z}$ are the same. Hence we have the relations

$$\begin{aligned}L^2(\chi_\perp\rho^2+\mu)&=(\chi_\perp-\chi_\parallel)/\gamma_*{}^2L^2,\\ \rho_1L^2\chi_\parallel-\rho(\chi_\perp-\chi_\parallel)&=(\chi_\perp-\chi_\parallel)/\gamma_*L^2,\end{aligned} \tag{44}$$

which connect ρ, ρ_1, and μ with the magnetomechanical ratio γ_* for a spin wave.

The AFMR spectrum is given by Eqs. (42). It is here important that the second derivatives in them give the same contribution as the first derivatives. Using (44) we get from the calculation the simple formula

$$\omega_{1,2}=\gamma_*(H_0\pm H), \tag{45}$$

where H_0 is exactly the same thermodynamic spin-flip field. This formula is the same as Eq. (25) from the Onsager version with

$$\gamma_\parallel=\gamma_*+O(\exp(-T_c/T)). \tag{46}$$

On the other hand, neither Eq. (25) with $\gamma_\parallel\neq\gamma_*$, nor Kittel and Keffer's Eq. (28) give the correct low-temperature asymptotic behavior (45) which, however, is the same as the result from microscopic theories (cf.[14]).

In all other variants of field orientations and signs the equations of motion simplify even more. In fact, if we neglect terms which are of higher accuracy (cf. the preceding section) we can use for finding the frequency spectrum the Lagrangian function

$$\mathscr{L}=\rho L\left[\mathbf{l}\times\frac{d\mathbf{m}}{dt}\right]+\frac{1}{2}\mu L^2\left(\frac{d\mathbf{l}}{dl}\right)^2-i. \tag{47}$$

Calculations give for the AFMR spectrum the same Eqs. (29), (31), and (32) where, however,

$$\gamma_\perp=\gamma_*+O(\exp(-T/T)). \tag{48}$$

The structure of all low-temperature formulae obtained in this way is the same as that given by microscopic or phenomenological theories (cf.[4,5,14]). However, they allow us to find the temperature dependence of the AFMR frequencies more exactly and in a more economical way; this occurs in the spectra only through the spin-flip field $H_0(T)$ and $\gamma_*(T)$.

In particular, in the Holstein–Primakoff approximation, i.e., in the large spin S approximation, $\gamma_*(T)$ can be found immediately if we use Oguchi's results.[14] In his paper the energy of the spin wave system, as in Landau's Fermi liquid theory ([18], see also[12], Sec. 68) is determined in the form of a functional of the spin wave distribution function $n_{1,2}$ for the two kinds of oscillations in (45). In Oguchi's theory the functional has the form

$$E\{n\}=\sum_{\mathbf{k}}(\varepsilon_{1\mathbf{k}}n_{1\mathbf{k}}+\varepsilon_{2\mathbf{k}}n_{2\mathbf{k}})+\frac{1}{2}\sum_{\mathbf{k}\mathbf{k}'}f^{11}_{\mathbf{k}\mathbf{k}'}n_{1\mathbf{k}}n_{1\mathbf{k}'}+\sum_{\mathbf{k}\mathbf{k}'}f^{12}_{\mathbf{k}\mathbf{k}'}n_{1\mathbf{k}}n_{2\mathbf{k}'}+\frac{1}{2}\sum_{\mathbf{k}\mathbf{k}'}f^{22}_{\mathbf{k}\mathbf{k}'}n_{2\mathbf{k}}n_{2\mathbf{k}'}+\dots,$$

where $\varepsilon_{1,2}$ are the spin wave energies when we neglect the interaction, while f^{11}, f^{12}, and f^{22} are the analogies of Landau's f-functions, which in Oguchi's theory are small: $f\sim 1/S$. Following Landau and defining the exact spin wave energy as

$$\tilde{\varepsilon}_{1,2}=\frac{\delta E}{\delta n_{1,2}}=\varepsilon_{1,2}+\sum_{\mathbf{k}'}f^{11,22}_{\mathbf{k}\mathbf{k}'}n_{1,2\mathbf{k}'}+\sum_{\mathbf{k}'}f^{12,21}_{\mathbf{k}\mathbf{k}'}n_{2,1\mathbf{k}'}+\dots$$

and substituting here

$$n_{1,2}=[\exp(\varepsilon_{1,2}/T)-1]^{-1},$$

we find after some straightforward calculations that

$$\gamma_*=\gamma_0(1-\chi_\parallel/2\chi_\perp),$$

where $\gamma_0=g\mu_0$ is the "free" magnetomechanical ratio, while $\chi_\parallel\sim T^2$, $\chi_\perp=\text{const}$.

However, the difference between the low-temperature phenomenology proposed here and theories[4,5,14] which are valid only in the limit $S\to\infty$ can in principle be made even more pronounced. Namely, we can drop the requirement that the magnetic moment of the excitation be connected through Eq. (7) with the angular momentum. This is completely possible in metals where $\chi_\parallel(T=0)$ may be finite while, on the other hand, it seems altogether improbable that theories operating with the Heisenberg exchange Hamiltonian (e.g., Oguchi's theory[14]) will give anything which is principally different in terms of order e^{-S}.

There are no special difficulties in performing calculations with the Lagrangian function (39) without the limitations (44). Their results consist in the fact that Eq. (46) is then violated while Eq. (48) remains. The AFMR frequencies will in that version of the theory thus again be described by Eqs. (25), (29), (31), and (32) where now, however,

$$\gamma_\parallel=\frac{\rho(\chi_\perp-\chi_\parallel)-\rho_1L^2\chi_\parallel}{L^2(\chi_\perp\rho^2+\mu)},\qquad(49)$$

$$\gamma_\perp^2=\gamma_*^2=\frac{\chi_\perp-\chi_\parallel}{L^2(\chi_\perp\rho^2+\mu)}.\qquad(50)$$

It would be interesting to observe the new qualitative features, viz. that $\gamma_\parallel\neq\gamma_\perp=\gamma_*$, which arise in such a theory experimentally. Of most interest would be the feature involving the presence at low temperatures of a term under the square root sign in Eq. (25) which is quadratic in the field H. We emphasize once again that this difference must be of the order of magnitude of the exchange in contrast to the trivial relativistic corrections which appear due to the dependence of H_0^2 on the external field.

5. $\mathrm{FeCO_3}$, $\mathrm{MnCO_3}$, $\mathrm{CoCO_3}$

For these substances we must add to the free energy, given by Eq. (15) a term $b[\mathbf{L}\times\mathbf{M}]_z$ corresponding to weak ferromagnetism.

a) $\mathrm{FeCO_3}$. *Weak field* $\mathbf{H}\parallel\mathbf{z}$. We add to the energy of the oscillations f (Eq. (17)) a term $b[\mathbf{l}\times\mathbf{m}]\mathbf{z}$. Equation (25) remains for the AFMR spectrum, in which we need only change the definition of the spin-flip field

$$H_0^2=\frac{a-b^2\chi_\perp}{\chi_\perp-\chi_\parallel}.\qquad(51)$$

b) $\mathrm{MnCO_3}$, $\mathrm{CoCO_3}$. $\mathbf{H}\parallel\mathbf{y}$, $\mathbf{L}\parallel\mathbf{x}$. These materials have a weak ferromagnetic moment M_0 which has the absolute magnitude $\chi_\perp|b|L$. In equilibrium in an external field we have instead of (16)

$$\mathbf{M}=b\chi_\perp[\mathbf{z}\times\mathbf{L}]+\chi_\perp\mathbf{H}=\mathbf{y}(M_0+\chi_\perp H).$$

Instead of Eq. (21) we shall now have for f

$$f=\frac{1}{2L^2\chi_\parallel}(\chi_\perp H+M_0)[(\chi_\perp-\chi_\parallel)H+M_0]l_y^2+\frac{1}{2}\left[|a|+\frac{1}{L^2\chi_\perp}M_0(\chi_\perp H+M_0)\right]l_z^2+\frac{1}{L\chi_\parallel}[(\chi_\perp-\chi_\parallel)H+M_0]l_ym_x$$
$$+{}^1/{}_2\chi_\parallel^{-1}m_x^2+{}^1/{}_2\chi_\perp^{-1}m_z^2+{}^1/{}_2Al_x^2+BHl_xm_y+{}^1/{}_2\chi_\perp^{-1}m_y^2.\qquad(52)$$

The equations of motion in the Onsager version also are changed. The first of the equations remains with the required accuracy in the old form:

$$\frac{d\mathbf{l}}{dt}=\gamma_1L\left[\mathbf{x}\times\frac{\partial f}{\partial\mathbf{m}}\right],\qquad(53')$$

while instead of the second one we must write

$$\frac{d\mathbf{m}}{dt}=\gamma_1L\left[\mathbf{x}\times\frac{\partial f}{\partial\mathbf{l}}\right]+(\gamma_5L^2\chi_\perp H+\alpha\gamma_1M_0)\left[\mathbf{y}\times\frac{\partial f}{\partial\mathbf{m}}\right].\qquad(53'')$$

The appearance of the term

$$\alpha\gamma_1M_0\left[\mathbf{y}\times\frac{\partial f}{\partial\mathbf{m}}\right]$$

with a new phenomenological coefficient α is connected with the fact that the symmetry of crystals which permit the appearance of the term $[\mathbf{L}\times\mathbf{M}]_z$ in the energy allows also the appearance of a term of the form

$$\left[[\mathbf{z}\times\mathbf{L}]\times\frac{\partial f}{\partial\mathbf{m}}\right]$$

in the equation of motion for $\mathbf{m}$. We remind ourselves that $\mathbf{M}_0\sim[\mathbf{z}\times\mathbf{L}]$, we are led to Eq. (53'').

We note also that the appearance of the coefficient α destroys the "easy plane" symmetry, introducing its own kind of dynamic anisotropy which exists even when there is no static anisotropy in the plane (Eq. (52)).

We give only the most interesting formula for the "ferromagnetic" mode ω_1:

$$\omega_1^2=H^2\gamma_\perp^2+\frac{\gamma_*^2M_0H}{\chi_\perp-\chi_\parallel}-2H\Delta(\gamma_\perp^2-\gamma_*^2)^{1/2}+\Delta^2,\qquad(54)$$

where the dynamic gap Δ is given by the formula

$$\Delta^2=\gamma_*^2(\alpha-1)^2M_0^2/\chi_\parallel(\chi_\perp-\chi_\parallel).$$

At low temperatures the solution of problem (47) with f from (52) gives for any version of the low-temperature theory (as in both $\gamma_*=\gamma_\perp$)

$$\omega_1^2/\gamma_*^2=H^2+M_0H/(\chi_\perp-\chi_\parallel). \tag{55}$$

There is thus no dynamic anisotropy in the spin wave region:

$$\alpha=1+O(\exp(-T_c/T)). \tag{56}$$

6. MnF$_2$, CoF$_2$

We consider here the easy-axis MnF$_2$, CoF$_2$ in a weak field parallel to the crystal axis. Their symmetry allows in the energy (15) a term $b(L_xM_y+L_yM_x)$. This leads to the additional analogous expression $b(l_xm_y+l_ym_x)$ in the expression (17) for the energy of the oscillations. In particular, the equation for the thermodynamic spin-flip field H_0 is changed. In fact, now

$$H_0=\left[\frac{(a-b^2\chi_\parallel)L^2}{\chi_\perp-\chi_\parallel}\right]^{1/2}-|b|L. \tag{57}$$

The evaluation of the spectra of the oscillations by means of Eqs. (24) or (42) leads at high temperatures to

$$\omega_{1,2}^2=\left\{\left[\frac{\gamma_*^2(a-b^2\chi_\perp\beta)L^2}{\chi_\perp-\chi_\parallel}+H^2(\gamma_\parallel^2-\gamma_*^2)\right]^{1/2}\pm H\gamma_\parallel\right\}^2-\frac{\gamma_*^4b^2L^2}{\gamma_\parallel^2}. \tag{58}$$

We have here for the sake of simplicity denoted by β the quantity

$$\beta=1-\frac{\gamma_*^2}{\gamma_\parallel^2}\left(1-\frac{\chi_\parallel}{\chi_\perp}\right). \tag{59}$$

The same formula is also retained in the second version of the spin wave theory with $\gamma_\parallel\neq\gamma_\perp=\gamma_*$. In the version with all gammas equal $\beta=\chi_\parallel/\chi_\perp$ and Eq. (58) simplifies:

$$\frac{\omega_{1,2}^2}{\gamma_*^2}=\left\{\left[\frac{(a-b^2\chi_\parallel)L^2}{\chi_\perp-\chi_\parallel}\right]^{1/2}\pm H\right\}^2-b^2L^2. \tag{60}$$

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[1] One can find a more detailed description how one must construct equations such as (1) in the already cited papers by Gufan[7] and by Andreev and Marchenko.[9]

[2] Here and in what follows the equilibrium values of the moments will be denoted by captials: **M**, **L**, and so on, and the deviations by lower case letters: **m**, **l**, and so on.

[3] As in the ferromagnetic case, the longitudinal mode is usually a relaxational mode. We shall not spend time on it and in the subsequent calculations we drop the index ⊥ of **m** and **l**.

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