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ENERGY OF MAGNETIC EXCITATIONS AND MAGNETIZATION IN COMPRESSIBLE FERROMAGNET

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ENERGY OF MAGNETIC EXCITATIONS AND MAGNETIZATION IN COMPRESSIBLE FERROMAGNET

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1. Introduction

A new approach to the theory of the spin-phonon interaction in the ferromagnetic crystals, based on the assumption that the exchange integral is a function of the instant disposition of the atoms, which change because of their thermal movements, has been proposed in ref. /1/. As compared to the conventional approach (see, e.g. ref /2/), this method is not restricted only to consideration of the linear terms in the expansion of the exchange integral, but allows one to consider in self-consistent manner the effects of the anharmonicity in lattice vibrations, which can be substantial in some definite cases.

This approach gives us a possibility to investigate in self-consistent manner the phonons and the magnetic excitations in the crystals /3/. On the basis of this method, using the theory of anharmonic crystals /4,5/, in ref. /6/ the equation of state of the ferromagnetic crystal has been derived, which makes it possible to consider in a unique way its thermal, magnetic and mechanical properties.

The purpose of the present work is to investigate the ferromagnetic crystal under a constant pressure and at low temperature, to find out the explicit expressions for the temperature dependence of the lattice constant and its contribution to the magnetization. We use the pseudo-harmonic approximation /4/ for the phonon system and the mean field approximation /7/ for the spin system, which at low temperatures gives us the correct temperature dependence of the magnetization. In ref. /8/ within the framework of the Ising model a similar approach was used to investigate the magnetization in the vicinity of the Curie point.

In Section 2 the Hamiltonian and the equation of state of the anharmonic ferromagnetic crystal and the magnetic excitation spectrum in the mean field approximation and at low temperatures are discussed. In Section 3 an analysis of the spectrum and the magnetization with the thermal expansion of the lattice taken into account is given.

2. Hamiltonian of the System. Green Function and Approximate Calculation of the Spectrum

Let us consider a ferromagnetic crystal, consisting of N magnetic atoms, which form a simple Bravais lattice. We assume that the spin-spin interaction can be described by the Heisenberg Hamiltonian and the interaction between the atoms determines the potential energy $U(\vec{R}_1 \ldots, \vec{R}_N)$. The total Hamiltonian of the system can be written in the form

$$\mathcal{H} = H_L + H_s + H_1 , \tag{1}$$

$$H_L = \sum_{\ell} \frac{\vec{p}_{\ell}^{\,2}}{2M} + U(\vec{R}_1, \ldots, \vec{R}_N), \tag{1a}$$

$$H_s = -\mu H \sum_{\ell} S_{\ell}^{z} - \frac{1}{2} \sum_{\ell, m} J(\vec{R}_{\ell} - \vec{R}_m)\, \vec{S}_{\ell} \cdot \vec{S}_m , \tag{1b}$$

$$H_1 = - \sum_{\ell, a} F_{\ell}^{a} R_{\ell}^{a} . \tag{1c}$$

$\vec{P}_{\ell}$ and $\vec{R}_{\ell}$ are the conjugate momentum and coordinate of the atom in the lattice site ℓ, μ is the Bohr magneton, H - the external magnetic field, $\vec{S}_{\ell}$ - the spin of the atom in the lattice site ℓ, $J(\vec{R}_{\ell} - \vec{R}_m)$ - the exchange integral, which we assume to be positive. H_1 describes the effect of the external forces $\vec{F}_{\ell}$, which deform the crystal lattice.

The equilibrium positions of the atoms in the lattice points ℓ_a are determined by the equality:

$$R_{\ell}^{a} = \langle R_{\ell}^{a} \rangle + u_{\ell}^{a} = \ell_a + u_{\ell}^{a} , \tag{2}$$

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The equilibrium positions of the atoms in the lattice points ℓ_α are determined by the equality:

$$R^\alpha_\ell = \langle R^\alpha_\ell \rangle + u^\alpha_\ell = \ell_\alpha + u^\alpha_\ell , \tag{2}$$

where the statistical average $\langle \dots \rangle$ is calculated at the equilibrium state of the crystal by the Hamiltonian [1]:

$$\langle \dots \rangle = Sp(e^{-\frac{\mathcal{H}}{T}} \dots)/Sp(e^{-\frac{\mathcal{H}}{T}}). \tag{3}$$

Applying the expansion of the potential energy in powers of u^α_ℓ - the thermal displacements of the atoms, we can write:

$$H_L = \sum_\ell \frac{\vec{p}^{\,2}_\ell}{2M} + \sum_{n=0}^{\infty} \frac{1}{n!} \sum_{1\dots n} \Phi_{1\dots n}\, u_1 \dots u_n , \tag{4}$$

where we introduce $u_1 = u^{\alpha_1}_{\ell_1}$ etc. and

$$\Phi_{1\dots n} \equiv \Phi^{\alpha_1\dots\alpha_n}_{\ell_1\dots\ell_n} = \nabla^{\alpha_1}_{\ell_1} \dots \nabla^{\alpha_n}_{\ell_n} U_0(\ell_1,\dots,\ell_n). \tag{5}$$

A similar expansion of the exchange integral in powers of u^α_ℓ gives us:

$$H_s = -\mu H \sum_\ell S^z_\ell - \frac{1}{2}\sum_{\ell,m}\sum_{n=0}^{\infty}\frac{1}{n!}\sum_{1\dots n} J_{1\dots n}(\vec{\ell}-\vec{m})\, u_1 \dots u_n\, \vec{S}_\ell \cdot \vec{S}_m \tag{6}$$

where

$$J_{1\dots n}(\vec{\ell}-\vec{m}) = J^{\alpha_1\dots\alpha_n}_{\ell_1\dots\ell_n}(\vec{\ell}-\vec{m}) = \nabla^{\alpha_1}_{\ell_1}\dots\nabla^{\alpha_n}_{\ell_n} J_0(\vec{\ell}-\vec{m}) =$$

$$= \prod_{i=1}^{n}(\delta_{in} - \delta_{im})\, \nabla^{\alpha_1}_{\ell}\dots\nabla^{\alpha_n}_{\ell} J_0(\vec{\ell}-\vec{m}). \tag{7}$$

To take into account the anharmonicity of the atom vibrations, we will keep every term in the expansion in (4) and (6) unlike the common harmonic approximation for the lattice Hamiltonian and the linear approximation for the spin-phonon interaction [2].

Taking into account that the average force acting on every atom is equal to zero and assuming the spin-phonon interaction to be small enough in the case of isotropic pressure p we get for the equation of state [6]:

$$p = -\frac{1}{3V}\sum_{\ell,a} \left\langle \frac{\partial}{\partial R^a_\ell} U \right\rangle + \frac{1}{6V}\sum_{\ell,m,a} (\vec{\ell}-\vec{m})^a_m \langle \vec{S}_\ell \cdot \vec{S}_m \rangle \times$$

$$\times \left\langle \frac{\partial}{\partial R^a_\ell} J(\vec{R}_\ell - \vec{R}_m) \right\rangle . \qquad (8)$$

This expression determines the dependence of the equilibrium lattice parameters on the temperature and the external forces.

To find the magnetic excitation spectrum we will use the Green function method and consider the two-time Green function composed by the spin operators $S^{\pm}_\ell = S^x_\ell \pm i S^y_\ell$ /9/:

$$G_{\ell\ell'}(t-t') = \langle\langle S^+_\ell(t) | S^-_{\ell'}(t') \rangle\rangle . \qquad (9)$$

Using the equations of motion for the operators in Heisenberg representation we get the following equation for the Green function (9):

$$\left(i\frac{\partial}{\partial t} - \mu H\right) G_{\ell\ell'}(t-t') = 2\langle S^z \rangle \delta_{\ell\ell'} \delta(t-t') + \qquad (10)$$

$$+ \sum_{n=0}^{\infty} \frac{1}{n!} \sum_{m,1\ldots n} J_{1\ldots n}(\vec{\ell}-\vec{m}) \langle\langle u_1 \ldots u_n (S^z_m S^+_\ell - S^z_\ell S^+_m)_{(t)} | S^-_{\ell'}(t') \rangle\rangle .$$

Taking into account the spin-phonon interaction in the lowest order only, we get a renormalization of the exchange integral in the mean phonon field and for the Green function in (10) we can write /1, 3, 10/:

$$\langle\langle u_1 \ldots u_n S^z_m S^z_\ell | S^-_{\ell'} \rangle\rangle \approx \langle u_1 \ldots u_n \rangle \langle\langle S^z_m S^+_\ell | S^-_{\ell'} \rangle\rangle . \qquad (11)$$

For the spin Green function we use also the mean field approximation /7/, which does not take into account the inelastic spin-spin interactions:

$$\langle\langle S^+_\ell S^z_m | S^-_{\ell'} \rangle\rangle = A_{\ell m} \langle\langle S^+_\ell | S^-_{\ell'} \rangle\rangle , \qquad (12)$$

where

$$A_{\ell m} = S + \frac{1}{2\langle S^z \rangle} \{ 2\langle (S^z_\ell - S)(S^z_m - S) \rangle - 2S(S - \langle S^z \rangle) + \qquad (13)$$

$$+ \langle S^-_\ell S^+_m \rangle \} .$$

After Fourrier transformation of the Green function according to:

$$G_{\ell\ell'}(t-t') = \frac{1}{2\pi}\int_{-\infty}^{\infty} \frac{1}{N}\sum_q e^{-i\vec{q}\cdot(\vec{\ell}-\vec{\ell}') - i\omega(t-t')} G_q(\omega) d\omega \qquad (14)$$

from (10) we get the following expression:

$$G_q(\omega) = \frac{2\langle S^z \rangle}{\omega - \mu H - E(\vec{q})} , \qquad (15)$$

$$E(\vec{q}) = S(\tilde{\tilde{J}}_0 - \tilde{\tilde{J}}_q) + \frac{1}{N}\sum_{q'} \phi(q')(\tilde{\tilde{J}}_{q'} - \tilde{\tilde{J}}_{q-q'}) , \qquad (16)$$

where

$$\phi(q) = \frac{1}{N\langle S^z \rangle} \sum_{\ell,m} e^{-i\vec{q}\cdot(\vec{\ell}-\vec{m})} \{ \langle (S^z_\ell - S)(S^z_m - S) \rangle +$$

$$+ \frac{1}{2}\langle S^-_\ell S^+_m \rangle - S(S - \langle S^z_\ell \rangle) \} \qquad (17)$$

and $\tilde{\tilde{J}}(q)$ is the Fourrier transform of the renormalized exchange integral:

$$\tilde{\tilde{J}}_{\ell m} = \langle J(\vec{R}_\ell - \vec{R}_m) \rangle \approx \sum_{n=0}^{\infty} \frac{1}{n!}\frac{1}{2} \{ \langle (u_\ell - u_m)^a (u_\ell - u_m)^\beta \rangle \times \qquad (18)$$

$$\times \nabla^a_\ell \nabla^\beta_\ell \} J_0(\vec{\ell}-\vec{m}) .$$

The approximate expression (18) represents the renormalized exchange integral in pseudoharmonical approximation /5/.

If we consider the low temperature limit of the spectrum (16), using the Holstein-Primakoff representation /11/.

$$S^z_f = S - n_f , \qquad n_f = a^+_f a_f$$
$$\langle S^-_\ell S^+_m \rangle \approx 2S \langle a^+_\ell a_m \rangle (1 + O(n_f)) \tag{19}$$
$$[a_f , a^+_{f'}] = \delta_{ff'}$$

and keeping the terms up to the first order in excitations number N_q:

$$\langle a^+_\ell a_m \rangle = \frac{1}{N} \sum_q e^{i\vec{q}\cdot(\vec{\ell}-\vec{m})} N_q$$

we obtain for the spectrum (16) the expressions:

$$E(\vec{q}) = S(\tilde{\tilde{J}}_0 - \tilde{\tilde{J}}_q) - \frac{1}{N} \sum_{q'} N_{q'} (\tilde{\tilde{J}}_0 + \tilde{\tilde{J}}_{q-q'} - \tilde{\tilde{J}}_q - \tilde{\tilde{J}}_{q'}) . \tag{20}$$

This expression coincides with Dyson's expression /12/, but instead of J_q here $\tilde{\tilde{J}}_q$ appears because of the anharmonicity of the lattice vibrations and the spin-phonon interaction.

3. Analysis of the Spectrum and Magnetization Taking into Account the Thermal Expansion of the Lattice

Here and throughout this part we shall suppose that the lattice potential can be described by two-body interactions:

$$U(\vec{R}_1,\ldots,\vec{R}_N) = \frac{1}{2} \sum_{n,m} \phi(\vec{R}_m - \vec{R}_n) \tag{21}$$

and we shall consider the spin and the phonon systems in the nearest neighbour approximation. Under these assumptions the equation of state (8) takes the form:

$$p = - \frac{z\ell}{6V} \{\tilde{\tilde{\phi}}'(\ell) - \tilde{\tilde{J}}'(\ell) \langle \vec{S}_\ell . \vec{S}_0 \rangle \} , \tag{22}$$

where z is the number of the nearest neighbours, ℓ is the separation between the nearest neighbours, V is the volume of the elementary cell, the prime implies differentiation with respect to ℓ.

The separation between the nearest neighbours under a constant pressure depends on the temperature and the equation of state at zero pressure takes the form:

$$\tilde{\tilde{\phi}}'(\ell) - \langle \vec{S}_\ell . \vec{S}_0 \rangle \tilde{\tilde{J}}'(\ell) = 0 . \tag{23}$$

The renormalized pair potential and exchange integral in pseudoharmonic approximation /5/ can be represented like this:

$$\tilde{\tilde{\phi}}(\ell) = \sum_{n=0}^{\infty} \frac{1}{n!} \left(\frac{\bar{u}^2}{2}\right)^n \phi^{(2n)}(\ell),$$
$$\tilde{\tilde{J}}(\ell) = \sum_{n=0}^{\infty} \frac{1}{n!} \left(\frac{\bar{u}^2}{2}\right)^n J^{(2n)}(\ell), \tag{24}$$

where $\bar{u}^2 \equiv \bar{u}^2_\ell = \langle [\vec{\ell}.(\vec{u}_\ell - \vec{u}_0)]^2 \rangle / \ell^2$. At low temperatures we have $\bar{u}^2 \equiv \bar{u}^2_T = \bar{u}^2_{T=0} + \xi$, where $\xi \approx T^4$ (see, e.g. ref /5/). Then (24) takes the form:

$$\tilde{\tilde{\phi}}(\ell) \approx \tilde{\tilde{\phi}}_0(\ell) + \xi \tilde{\tilde{\phi}}''_0(\ell),$$
$$\tilde{\tilde{J}}(\ell) \approx \tilde{\tilde{J}}_0(\ell) + \xi \tilde{\tilde{J}}''_0(\ell), \tag{24a}$$

where the expressions for $\tilde{\tilde{\phi}}_0(\ell)$ and $\tilde{\tilde{J}}_0(\ell)$ can be obtained from (24) by means of substitution $\bar{u}^2 = \bar{u}^2_{T=0}$.

The separation between the nearest neighbours ℓ can be expressed like $\ell \equiv \ell(T) = \ell_0 + \delta\ell(T)$, where ℓ_0 is the equilibrium separation at zero pressure and zero temperature, determined by the equation:

$$\tilde{\tilde{\phi}}'_0(\ell_0) - S^2 \tilde{\tilde{J}}'_0(\ell_0) = 0 . \tag{25}$$

Using (24a) and taking the terms up to T^4, from the equation of state (23) we get:

$$\delta\ell(T)\{\tilde{\tilde{\phi}}''_0(\ell_0) - S^2\tilde{\tilde{J}}''_0(\ell_0)\} = (\langle \vec{S}_\ell . \vec{S}_0 \rangle - S^2)\tilde{\tilde{J}}'_0(\ell_0) + \xi\{S^2\tilde{\tilde{J}}'''_0(\ell_0) - \tilde{\tilde{\phi}}'''_0(\ell_0)\}. \tag{26}$$

Using the results of ref. /5/ and (19) from (26) we obtain:

$$\delta\ell(T) = AT^{\frac{5}{2}} + BT^{4}, \tag{27}$$

where for simple cubic lattice:

$$A = -2\pi S Z_{5/2}\left(\frac{\mu H}{T}\right)\tilde{\tilde{J}}'_{0}(\ell_0)/[4\pi S\tilde{\tilde{J}}_0(\ell_0)]^{5/2} f(\ell_0)$$

$$B = \pi^4 [S^2 \tilde{\tilde{J}}'''_0(\ell_0) - \tilde{\tilde{\phi}}'''_0(\ell_0)]/10\omega_D^3 \, \phi''(\ell_0) f(\ell_0)$$

$$f(\ell_0) = \tilde{\tilde{\phi}}''_0(\ell_0) - S^2 \tilde{\tilde{J}}''_0(\ell_0), \quad Z_p(x) = \sum_{n=1}^{\infty} n^{-p} e^{-nx},$$

$$\omega_D = 1{,}05\sqrt{8\phi''(\ell_0)/M}.$$

In the expression (27) for the thermal expansion of the lattice constant besides the usual term BT^4 a supplementary term $AT^{5/2}$ appears because of the spin-phonon interaction. Obviously in the low temperature region ($\theta = T/4\pi S\tilde{\tilde{J}}_0(\ell_0) \ll 1$) this term is the dominating one. In the considered case for ferromagnet we have $A > 0$ and $B>0$. This is a consequence of the fact that usually $\tilde{J}'_0(\ell_0) < 0$, $f(\ell_0) > 0$ (because $\tilde{\phi}''_0(\ell_0) > 0$ and $\tilde{\phi}''_0(\ell_0) > S^2 \tilde{J}''_0(\ell_0)$ and $\tilde{\phi}'''_0(\ell_0) < 0$.

From (27) for the thermal expansion coefficient we get:

$$\eta = \frac{1}{\ell_0}\frac{d}{dT}\delta\ell(T) = \frac{1}{\ell_0}\left(\frac{5}{2}AT^{3/2} + 4BT^3\right). \tag{28}$$

To obtain the expression for the relative magnetization at low temperature

$$\sigma = 1 - \frac{1}{SN}\sum_q \left[\exp\frac{\mu H + E(\vec{q})}{T} - 1\right]^{-1} \tag{29}$$

we have to calculate the spectrum (20) which in the nearest neighbours approximation for the simple cubic lattice when $q \to 0$ and $T \to 0$ takes the form:

$$E(\vec{q}) = S^2 q^2 \ell_0^2 \tilde{\tilde{J}}_0(\ell_0)\left[1 + \xi\frac{\tilde{\tilde{J}}''_0(\ell_0)}{\tilde{\tilde{J}}_0(\ell_0)} + \delta\ell(T)\frac{\tilde{\tilde{J}}'_0(\ell_0)}{\tilde{\tilde{J}}_0(\ell_0)}\right] \times$$

$$\times\left[1 - \pi\frac{Q(S)}{S}\left(\frac{T}{4\pi S\tilde{\tilde{J}}_0(\ell_0)}\right)^{5/2} Z_{5/2}\left(\frac{\mu H}{T}\right)\right], \tag{30}$$

where $Q(S) = (1 + 0{,}03S^{-1})/(1 - 0{,}1S^{-1}) + 0{,}17S^{-1}$.

The second term in the last brackets in (30) is caused by the integral term in (20) (see, e.g. (18) in ref. /13/). Using (27) and (30) for the relative magnetization (29) we obtain:

$$\sigma = 1 - a\theta^{3/2} - \beta\theta^4 - \gamma\theta^4, \tag{31}$$

where we introduce:

$$a = \frac{1}{S} Z_{3/2}\left(\frac{\mu H}{T}\right)$$

$$\beta = \frac{3}{2}\pi\frac{Q(S)}{S} Z_{3/2}\left(\frac{\mu H}{T}\right) Z_{5/2}\left(\frac{\mu H}{T}\right)$$

$$\gamma = 3\pi Z_{3/2}\left(\frac{\mu H}{T}\right) Z_{5/2}\left(\frac{\mu H}{T}\right)[\tilde{\tilde{J}}'_0(\ell_0)]^2 / \tilde{\tilde{J}}_0(\ell_0) f(\ell_0).$$

In the expression (31) besides the Bloch term ($a\theta^{3/2}$) and the Dyson term ($\beta\theta^4$) there appears a third term ($\gamma\theta^4$) due to the dependence of the lattice constant on the temperature.

4. Discussion

The principal result of the present paper is the microscopic derivation of the thermal expansion of the lattice constant, where the coefficients are explicitly expressed by the spin-phonon interaction constant. The temperature dependence in (27) coincides with the result obtained in the phenomenological way in ref. /2/. There the term proportional to $T^{5/2}$ appears not from taking into account the thermal expansion of the lattice constant, but from the renormalization of the magnetic excitation spectrum.

The expression (27) indicates that at low temperatures $(\theta = T/4\pi S\tilde{J}_0(\ell_0) \ll 1)$ the spin system exerts an essential influence on the thermal expansion. The first term of (27) introduces in the magnetization an addition proportional to T^4, the coefficient γ of which contains the factor $[\tilde{J}'_0(\ell_0)]^2$ playing in our case the role of the effective coupling constant.

The influence of the thermal expansion on the magnetization at low temperatures can be comparable with the spin-spin interaction effect. It is seen from (31) that $\gamma \approx \beta$ if the condition $[\tilde{J}'_0(\ell_0)]^2 \approx \tilde{J}_0(\ell_0) f(\ell_0)$ is fulfilled, i.e. if the spin-phonon interaction is strong enough.

We point out that the method of the present paper can be used in the investigations of the solid ^3He which represents a nuclear antiferromagnet and at the same time a strongly anharmonic crystal.

Analogous investigations for the temperatures in the vicinity of the Curie point will be carried out elsewhere.

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