at field $H=H_c=6.3$ kOe in a domain where, for $H \ll H_c$, $L \parallel H$; and to a continuous rotation of L in domains where L was oriented at angle 60° to H. In strong magnetic fields, $H \gg H_c$, independently of the orientation of H in the basal plane, $L \perp H$. On change of the orientation of the antiferromagnetic L in the basal plane of the crystal, for certain positions of L in this plane there appears a ferromagnetic moment σ_z along the c axis; its variation can be described with sufficient accuracy by the expression $\sigma_z=\sigma_z^0 \sin 3\psi$. In this work we have determined the values of the effective fields responsible for the magnetic properties of this monocrystal.

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[1]M. Melamud, J. Makovsky, H. Shaked, and S. Shtrikman, Phys. Rev. **B3**, 821 (1971).
[2]A. N. Bazhan and N. M. Kreĭnes, Pis'ma Zh. Eksp. Teor. Fiz. **15**, 533 (1972) [JETP Lett. **15**, 377 (1972)].
[3]A. N. Bazhan, Zh. Eksp. Teor. Fiz. **67**, 1520 (1974) [Sov. Phys. JETP **40**, 757 (1974)].
[4]P. J. Flanders, AIP Conf. Proc. 5, Part 2, 795 (1972).
[5]N. V. Fedoseeva, Pis'ma Zh. Eksp. Teor. Fiz. **21**, 108 (1975) [JETP Lett. **21**, 48 (1975)].
[6]A. N. Bazhan, A. S. Borovik-Romanov, and N. M. Kreĭnes, Prib. Tekh. Eksp. **16**, No. 1, 213 (1973) [Instrum. Exp. Tech. **16**, No. 1, 213 (1973) [Instrum. Exp. Tech. **16**, No. 1 261 (1973)].
[7]A. S. Borovik-Romanov, Itogi nauki, Antiferromagnetizm (Results of Science, Antiferromagnetism), Izd. Akad. Nauk SSSR, 1962.
[8]A. N. Andrienko and L. A. Prozorova, Zh. Eksp. Teor. Fiz. **74**, 1527 (1978) [Sov. Phys. JETP **47**, 798 (1978)].

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Spin–phonon mechanism of magnetic ordering in paramagnetic crystals

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The quantum field theory methods are applied to a phase transition in a system of paramagnetic ions interacting with acoustic phonons. It is shown that when a threshold condition, imposed on an external static magnetic field and paramagnetic ion density, is exceeded, a paramagnetic crystal has a critical temperature T_c below which it experiences spontaneous static deformation accompanied by a spontaneous magnetic ordering of the $\langle S_x S_z \rangle \neq 0$ type. The resonant response of the spin–phonon system to an external rf field and to sound is determined in the range $T \geq T_c$ and one of the resonance modes is found to be soft. The phase transition in question is not exhibited by a crystal with a macroscopically inhomogeneous distribution of paramagnetic ions.

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INTRODUCTION

Investigations of a paramagnetic system interacting with a radiation field in a resonator have shown that a phase transition may occur when the density of paramagnetic ions and an external static magnetic field exceed certain critical values.[1] The ordered thermodynamic phase is characterized by a spontaneous magnetic order directed at right-angles to the external field and by a static component of the radiation field.

It is of interest to consider an analogous problem of a phase transition in a spin–phonon system. The new thermodynamic phase should be manifested by magnetic

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ordering and thermal displacements of atoms in a crystal from the positions they have occupied in the high-symmetry phase. In other words, such a phase transition should give rise to a simultaneous static deformation of a paramagnetic crystal. We shall solve this problem by considering the temperature spectrum of collective excitations of the spin–phonon system and phonon Green functions. We shall consider the spin–phonon interaction within the framework of the generally accepted Van Vleck mechanism, which is the modulation of the crystal field by the lattice vibrations.[2]

A phase transition in a paramagnetic crystal can be analyzed phenomenologically by calculating the thermodynamic potential of paramagnetic ions interacting with static deformations of the crystal.[3]

1. INITIAL HAMILTONIAN

The explicit form of the spin–phonon interaction for $S = 1/2$ can be written as follows[2,4]:

$$V = S_\alpha H_\beta G'_{\alpha\beta\gamma\delta}\varepsilon_{\gamma\delta}, \tag{1}$$

and for $S > 1/2$, the interaction is a quadratic function of the spin operators:

$$V = \frac{1}{2}(S_\alpha S_\beta + S_\beta S_\alpha) G_{\alpha\beta\gamma\delta}\varepsilon_{\gamma\delta}, \quad \varepsilon_{\gamma\delta} = \frac{1}{2}\left(\frac{\partial u_\gamma}{\partial x_\delta} + \frac{\partial u_\delta}{\partial x_\gamma}\right). \tag{2}$$

Here, S_α are the spin components; α, β, γ, $\delta = x, y, z$; $G_{\alpha\beta\gamma\delta}$ is a fourth-rank tensor which governs the spin–phonon interaction; u_α are the components of the displacement vector. The selection of this type of the spin–phonon interaction is dictated mainly by the fact that it has been investigated experimentally quite thoroughly.[5] The interaction (2) usually predominates for paramagnetic ions with spin exceeding 1/2.

In the case of crystals with the cubic lattice the interaction (2) includes only two spin–phonon interaction constants and is of the form[5]

$$\begin{aligned} V = {} & G_{11}[{}^1/_4(2\varepsilon_{zz}-\varepsilon_{xx}-\varepsilon_{yy})(2S_z^2-S_x^2-S_y^2) \\ & +{}^3/_4(\varepsilon_{xx}-\varepsilon_{yy})(S_x^2-S_y^2)] + G_{44}[\varepsilon_{yz}(S_yS_z+S_zS_y) \\ & +\varepsilon_{xz}(S_xS_z+S_zS_x)+\varepsilon_{xy}(S_yS_x+S_xS_y)], \end{aligned} \tag{3}$$

where the displacement tensor expressed in the phonon operator representation is

$$\varepsilon_{\alpha\beta} = i\left(\frac{\hbar}{2mN}\right)^{1/2}\sum_{\mathbf{k},s}\frac{e_\alpha^{(s)}k_\beta}{[\Omega_s(k)]^{1/2}}(b_{\mathbf{k}s}e^{i\mathbf{kr}} - b_{\mathbf{k}s}^+ e^{-i\mathbf{kr}}). \tag{4}$$

Here, m is the mass of a unit cell in a crystal; N is the number of atoms in the crystal; α and β are the indices referring to the crystallographic axes; $e_\alpha^{(s)}$ are the unit vectors of the s-th polarization of the vibrations; $\Omega_s(\mathbf{k})$ is the vibration frequency; $b_{\mathbf{k}s}^+$ and $b_{\mathbf{k}s}$ are the phonon creation and annihilation operators; the wave vector $\mathbf{k}$ assumes only three directions along the principal crystallographic axes.

The total Hamiltonian of M paramagnetic ions interacting with phonons will be written in the form $H = \bar{H}_0 + V$, where

$$\bar{H}_0 = \hbar\omega_0\sum_{i=1}^{M} S_{zi} + \sum_{\mathbf{k},s}\hbar\Omega_s(\mathbf{k})\, b_{\mathbf{k}s}^+ b_{\mathbf{k}s}, \tag{5}$$

$\hbar\omega_0 = g_\parallel\mu_B H_0$; $g_\parallel$ is the longitudinal g factor; μ_B is the Bohr magneton; H_0 is an external static magnetic field parallel to the axis. Without loss of generality, we shall assume that $G_{11} \ll G_{44}$. Then, substituting Eq. (4) into Eq. (3) and retaining only the resonance terms, we find that the spin–phonon interaction operator is

$$\begin{aligned} V = {} & \frac{2\lambda_1}{M^{1/2}}\sum_{j=1}^{M} S_{+j}^2[b_1\exp(iqx_j) + b_2\exp(iqy_j)] \\ & + \frac{\lambda_2\sqrt{2}}{M^{1/2}}\sum_{j=1}^{M}\gamma_{+j}[b_3\exp(ikx_j) + b_4\exp(ikz_j) \\ & - ib_5\exp(iky_j) - ib_6\exp(ikz_j)] + \text{H.c.} \end{aligned} \tag{6}$$

The first term in Eq. (6) describes the processes of emission and absorption of acoustic phonons whose wave vector is q and frequency $\Omega_1(q) \approx 2\omega_0$; the second describes the corresponding processes for phonons of f frequency $\Omega_2(k) \approx \omega_0$ and the wave vector k;

$$\lambda_1 = (G_{44}q/2)(\hbar p/2m\Omega_1)^{1/2}, \quad \lambda_2 = (G_{44}k/2)(\hbar p/m\Omega_2)^{1/2}, \tag{7}$$

$$S_\pm = S_x \pm iS_y, \quad \gamma_\pm = S_\pm S_z, \quad p = M/N. \tag{8}$$

For simplicity, we shall at this stage consider the interaction of spins only with six phonon modes. Generalization to the case of an arbitrary number of phonon modes presents no difficulty and will be made later.

In the fermion representation the Hamiltonian of the spin–phonon system for $S = 1$ becomes

$$\bar{H}_0 = \sum_{j=1}^{M}\hbar\omega_0(a_{1j}^+a_{1j} - a_{-1j}^+a_{-1j}) + \sum_{\nu=1}^{6}\hbar\omega_\nu b_\nu^+ b_\nu, \tag{9}$$

$$\begin{aligned} V = {} & \sum_{\nu=1,2}\frac{\lambda_1}{M^{1/2}}\sum_{j=1}^{M}(a_{1j}^+a_{-1j}b_\nu\exp(i\mathbf{q}_\nu\mathbf{r}_j) \\ & + \frac{\lambda_2}{M^{1/2}}\sum_{j=1}^{M}\sum_{\nu=3}^{6}(a_{1j}^+a_{0j} + a_{0j}^+a_{-1j})\, b_\nu\exp(i\mathbf{k}_\nu\mathbf{r}_j) + \text{H.c.}, \end{aligned} \tag{10}$$

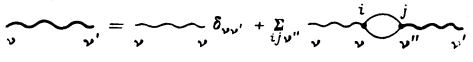
where the directions of the wave vectors are easily deduced from Eq. (6); the indices 1, 0, and −1 refer to the corresponding energy levels of paramagnetic ions in an external magnetic field.

2. PHONON GREEN FUNCTION

The existence of a phase transition in our system can be deduced from the temperature features of the phonon Green functions in the same way as in the case of second-order phase transitions in a Bose liquid.[6] Expanding the Green phonon functions

$$F_{\nu\nu'} = \langle T\{b_\nu(\tau)b_{\nu'}^+(0)\}\rangle$$

as a series in terms of the interaction (10), we find that the usual rules of the temperature diagram technique[7] give



Here, thin wavy lines represent zeroth phonon Green functions, which—according to Eq. (9)—are given by

$$F_{\nu\nu}^{0}(\omega_m)=-\frac{1}{i\omega_m-\omega_\nu},\quad \omega_m=\frac{2\pi m}{\beta},\quad m=0,\pm1,\pm2,\ldots\quad \beta=\frac{1}{kT}, \tag{11}$$

and the thick wavy functions represent the total phonon Green functions. The summation with respect to i and j is carried out over the crystal lattice sites occupied by paramagnetic ions. The Greek symbols are used for the phonon mode numbers. The thin curves in Eq. (1) are the zeroth fermions propagators, which—according to Eq. (9)—are

$$C_s^{0}(\omega_m)=-1/(i\omega_n-\varepsilon_s-\mu),\quad \omega=\pi(2n+1)/\beta,\quad n=0,\pm1,\pm2,\ldots, \tag{12}$$

where s is the number of an energy level of a paramagnetic ion; $\epsilon_\pm=\pm\omega_0$, $\epsilon_0=0$; μ is the chemical potential introduced to avoid the contribution of nonphysical states in the fermion representation of the spin operators by means of the projection operator[8,9]

$$\hat{P}=\frac{1}{\mathrm{Sp}\{\exp(-\beta\hat{H}_0)\}}\lim_{\mu\to\infty}e^{\beta\mu}. \tag{13}$$

As pointed out earlier,[10] in the thermodynamic limit N, $V\to\infty$ and $N/V=\mathrm{const}$, the only important vertices are the bare ones. Therefore, Eq. (A) does not include the polarization diagrams of the type

For this reason in the disordered temperature range the zeroth fermion Green functions (12) are exact because the mass operators for the finite-mode case vanish in the thermodynamic limit.

Everywhere, unless otherwise stated, we shall use the formula

$$\Delta(\mathbf{k}-\mathbf{k}')=\frac{1}{M}\sum_j\exp\{i(\mathbf{k}-\mathbf{k}')\mathbf{r}_j\}\approx\delta_{\mathbf{k},\mathbf{k}'}, \tag{14}$$

which is valid for a macroscopically homogeneous distribution of paramagnetic ions in a crystal with low $p\ll1$ and very high $p\sim1$ densities.[7]

The solution of Eq. (A) for $\nu=1$ and 2 has the form

$$F_{\nu\nu}=[(F_{\nu\nu}^{0})^{-1}-\lambda_1^{2}\Pi_{+-}]^{-1}; \tag{15a}$$

for $\nu=3$ and 5:

$$F_{\nu\nu}=[(F_{\nu\nu}^{0})^{-1}-\lambda_2^{2}(\Pi_{+0}+\Pi_{0-})]^{-1}; \tag{15b}$$

for $\nu=4$ and 6:

$$F_{44}=F_{44}^{0}[1-\lambda_2^{2}F_{66}^{0}(\Pi_{+0}+\Pi_{0-})]/\Delta,$$
$$F_{66}=F_{66}^{0}[1-\lambda_2^{2}F_{44}^{0}(\Pi_{+0}+\Pi_{0-})]/\Delta; \tag{15c}$$

$$\Delta=[1-\lambda_2^{2}F_{44}^{0}(\Pi_{+0}+\Pi_{0-})][1-\lambda_2^{2}F_{66}^{0}(\Pi_{+0}+\Pi_{0-})]-\lambda_2^{4}F_{44}^{0}F_{66}^{0}(\Pi_{+0}+\Pi_{0-})^{2}, \tag{15d}$$

where

$$\Pi_{ss'}=-\frac{1}{\beta}\sum_n G_s^{0}(\omega_n+\omega_m)G_{s'}^{0}(\omega_n). \tag{16}$$

Substituting in Eq. (16) the fermion Green functions (12), we find from the projection operator (13) that

$$\Pi_{+-}=Q(\beta)/(i\omega_m-2\omega_0),\quad \Pi_{+0}+\Pi_{0-}=-Q(\beta)/(i\omega_m-\omega_0), \tag{17}$$

where

$$Q(\beta)=\frac{2\,\mathrm{sh}\,\beta\omega_0}{1+2\mathrm{ch}\,\beta\omega_0}\quad(\hbar=1). \tag{18}$$

The termperature at which the denominator of any ν-th phonon Green function vanishes at $\omega_m=0$ determines the phase transition temperature.[6] Substituting the expressions (17) and (18) into (15), we find that for $\omega_m=0$ there are simultaneous singularities in the phonon Green functions of the fourth and sixth branches at a temperature β_c, given by the equation

$$Q(\beta_c)=\omega_0\omega_4/2\lambda_2^{2}. \tag{19}$$

It can be seen from Eq. (18) that this equation has a solution if the following threshold condition is satisfied

$$2\lambda_2^{2}>\omega_0\omega_4. \tag{20}$$

Since the average number of phonons for $\nu=4$ or 6

$$\langle b_\nu^{+}b_\nu\rangle=\sum_m F_{\nu\nu}(\omega_m)e^{i\omega_m(\tau\to-0)}$$

diverges in the limit $\beta\to\beta_c-0$, this phase transition represents the Bose–Einstein condensation.

There is no point in writing down, by analogy with Eq. (19), the equations for the Bose–Einstein condensation temperature of phonons of other branches because for $\beta>\beta_c$ the phonon Green functions assume a more complex form compared with Eq. (15) due to the appearance of the order parameter $\langle b_\mu/N^{1/2}\rangle$, where $\nu=4$ or 6.[7]

3. DYNAMIC SUSCEPTIBILITY OF A SPIN-PHONON SYSTEM AT $T\geqslant T_c$. SOFT MODE

The interaction of a spin system with an external rf probe field or with external hypersound is described by

$$\sum_j(S_{+j}h+S_{-j}h^{*}). \tag{21}$$

The reaction of the system to these perturbations is given by an analytic continuation of the polarization Green function[7]

$$\chi(\omega_m)=\sum_{i,j}\frac{1}{\beta}\int_{-\beta}^{\beta}\exp(-i\omega_m\tau)\,d\tau\langle T\{S_{+i}(\tau)S_{-i}(0)\}\rangle$$
$$=2\sum_{i,j}\sum_{a=1}^{4}\chi_{ij}^{(a)}(\omega_m),$$

where

$$\chi^{(1)}=\langle T(a_{1i}^{+}a_{0i}a_{0j}^{+}a_{1j})\rangle,\quad \chi^{(2)}=\langle T(a_{1i}^{+}a_{0i}a_{-j}^{+}a_{0j})\rangle,$$
$$\chi^{(3)}=\langle T(a_{0i}^{+}a_{-i}a_{-j}^{+}a_{0j})\rangle,\quad \chi^{(4)}=\langle T(a_{0i}^{+}a_{-i}a_{0j}^{+}a_{+j})\rangle.$$

Expanding these Green functions in terms of the polarization diagrams within the framework of the model of Eqs. (9) and (10), we obtain

$$\chi^{(1)}(\mathbf{k})=\Pi_{+0}/N+\lambda_2^2[\chi^{(1)}(\mathbf{k})+\chi^{(3)}(\mathbf{k})]F_{33}{}^0\Pi_{+0},$$
$$\chi^{(2)}(\mathbf{k})=\lambda_2^2[\chi^{(2)}(\mathbf{k})+\chi^{(4)}(\mathbf{k})]F_{33}{}^0\Pi_{+0},$$
$$\chi^{(3)}(\mathbf{k})=\lambda_2^2[\chi^{(1)}(\mathbf{k})+\chi^{(3)}(\mathbf{k})]F_{33}{}^0\Pi_{0-},$$
$$\chi^{(4)}(\mathbf{k})=\Pi_{0-}/N+\lambda_2^2[\chi^{(2)}(\mathbf{k})+\chi^{(4)}(\mathbf{k})]F_{33}{}^0\Pi_{0-},$$

where

$$\chi^{(\alpha)}(\mathbf{k})=\frac{1}{N}\sum_i \exp(i\mathbf{k}\mathbf{r}_{ij})\chi_{ij}^{(\alpha)}.$$

We have allowed here for the circumstance that analytic expressions for zeroth phonon Green functions $F^0_{\nu\nu}$ for $\nu=3$, 4, 5, and 6 can be regarded as identical in the resonance case $\omega_\nu \approx \omega_0$. Consequently, for the perturbation of the (21) type, we obtain the following resonance frequencies:

$$\Omega_1=\omega_0,\quad \Omega_{2,3}=\frac{\omega+\omega_0}{2}\pm\left[\left(\frac{\omega-\omega_0}{2}\right)^2+\lambda_2^2Q(\beta)\right]^{1/2}, \tag{22}$$

where ω is the frequency of the acoustic branches $\nu=3$, 4, 5, or 6.

Next, we can see from Eq. (6) that the interaction of the spin subsystem with an external probing hypersound can be written in the form

$$\sum_j (S_{+j}{}^2\xi+S_{-j}{}^2\xi^*) \tag{23}$$

or

$$\sum_j (\gamma_{+j}\eta+\gamma_{-j}\eta^*), \tag{24}$$

where ξ and η are the amplitudes of the probing sound. The case (23) gives the following resonance frequencies

$$\Omega_4=2\omega_0,\quad \Omega_{5,6}=\frac{\omega_1+2\omega_0}{2}\pm\left[\left(\frac{\omega_1-2\omega_0}{2}\right)^2+\lambda_1^2Q(\beta)\right]^{1/2} \tag{25}$$

and it is basically similar to the perturbation (21). A perturbation of the (24) type is of much greater interest. Such hypersound is in resonance at the following frequencies:

$$\Omega_7=\omega_0,\quad \Omega_8=\omega,\quad \Omega_{+-}=\frac{\omega+\omega_0}{2}\pm\left[\left(\frac{\omega-\omega_0}{2}\right)^2+2\lambda_2^2Q(\beta)\right]^{1/2}. \tag{26}$$

It follows from Eq. (19) that the branch of the collective excitations Ω_- of the spin–phonon system vanishes at the phase transition. This branch is a soft mode responsible for the spontaneous lowering of the high-temperature symmetry of the system.

4. DISCUSSION

In estimating the temperature of a phase transition we shall rewrite Eq. (19), subject to Eqs. (9) and (18), in the form

$$2\,\mathrm{sh}(\beta_c g_\parallel \mu_B H_0)/[1+2\,\mathrm{ch}(\beta_c g_\parallel \mu_B H_0)]=2mg_\parallel\mu_B H_0 v^2/pG_{44}{}^2 \tag{27}$$

and the threshold condition (22) in the form

$$g\mu_B H_0 < pG_{44}{}^2/2mv^2. \tag{28}$$

If the external magnetic field H_0 is sufficiently weak so that $\hbar\omega_0/kT_c < 1/2$, we readily find from Eq. (27) that

$$kT_c \approx {}^4/_3 pG_{44}{}^2/mv^2. \tag{29}$$

Thus, the temperature of a phase transition in the spin–phonon system is governed by the ratio of the square of the spin–phonon coupling constant to the kinetic energy of the propagation of sound per paramagnetic ion.

If we now turn to the experimental data,[11] we find that the highest spin–phonon coupling constant is exhibited by the Fe^{2+} paramagnetic ions in a CaF_2 crystal: $G_{44}=540$ cm^{-1}, $v=6.68\times10^5$ cm/sec, $m\approx1.3\times10^{-22}$ g. If we assume that the proportion of the paramagnetic ions to atoms in the whole crystal is $p=1/10$, we find from Eq. (28) that the threshold condition corresponds to $H_0<600$ Oe. It follows from Eq. (29) that the transition temperature is $T_c\approx0.04$°K. Bearing in mind that in fact the spin of a Fe^{2+} paramagnetic ion in CaF_2 is 2, we may expect an approximately fourfold increase in T_c for this paramagnetic crystal.[12]

We shall now discuss the results. If the threshold condition (28) is satisfied, a second-order phase transition may occur in a system of paramagnetic ions which interact resonantly with acoustic phonons. The ordered thermodynamic phase existing in the $T\leq T_c$ range is the Bose condensate of those phonons which are coupled most strongly to the paramagnetic ions. For this phase we have $\langle b_\nu/N^{1/2}\rangle$ and $\langle b_\nu{}^+/N^{1/2}\rangle$ which differ from zero and are c numbers.[13] Thus, according to Koloskova[4] a spontaneous static deformation is experienced by a paramagnetic crystal at such a phase transition. It is clear from Eq. (6) that the deformation is directed along the z axis. A phase transition also gives rise to a spontaneous magnetic polarization $\langle S_-S_z\rangle$, $\langle S_+S_z\rangle$. If $\lambda_1>\lambda_2$, the spontaneous deformation of a paramagnetic crystal is at right-angles to the z axis and the magnetic ordering is of the $\langle S_+^2\rangle$, $\langle S_-^2\rangle$ type.

Next, at T_c the frequency of response of the spin–phonon system to external acoustic vibration vanishes. Consequently, at a point $T=T_c$ the system becomes unstable in the presence of any, no matter how weak, homogeneous static deformation of the crystal. Thus, according to the theory of second-order phase transitions,[3] the isothermal compressibility of a paramagnetic crystal diverges in the limit $T\to T_c$.

We have considered the phase transition in the spin–phonon system solely in the resonance approach. In the case of a packet of phonon modes the equation (A) for the Green phonon functions can be rewritten in the form

$$F(\mathbf{k},\mathbf{k}',\omega_m)=F^0(\mathbf{k},m)\delta_{\mathbf{k},\mathbf{k}'}+F^0(\mathbf{k},m)\lambda_2(k)\Pi_{+-}(m)$$
$$\times\frac{1}{N}\sum_{\mathbf{k}''}\sum_{j=1}^{N}\lambda_2(\mathbf{k}'')F(\mathbf{k}'',\mathbf{k}',m)\exp[i(\mathbf{k}-\mathbf{k}'')\mathbf{r}_j]. \tag{30}$$

If we use Eq. (14), we find that the phase transition

temperature agrees with the previous result (29). However, if the distribution of paramagnetic ions in a crystal is macroscopically inhomogeneous or if the density is intermediate, $p<1$, the temperature of the phase transition may be broadened. To check this possibility, we shall first assume that the phase transition does occur in this case. Then, in the vicinity of the crystal temperature the phonon Green function $F(\mathbf{k}, \mathbf{k}', \omega_m=0)$ should be singular at $\mathbf{k}=\mathbf{k}'$. Hence, the solution of Eq. (30) can be written in the form

$$F(\mathbf{k},\mathbf{k}',0)=F^0(\mathbf{k},0)\delta_{\mathbf{k},\mathbf{k}'}+\frac{F^0(\mathbf{k},0)F^0(\mathbf{k}',0)\lambda_2(\mathbf{k})\lambda_2(\mathbf{k}')\Delta(\mathbf{k}-\mathbf{k}')}{1-\lambda_2(\mathbf{k}')\Pi_{+-}(0)\sum_{\mathbf{q}}F^0(\mathbf{q},0)\lambda_2(\mathbf{q})\Delta(\mathbf{k}-\mathbf{q})}. \quad (31)$$

Zero of the denominator on the right-hand side of Eq. (31) at $\mathbf{k}=\mathbf{k}'$ gives the phase transition temperature. If $\Delta(\mathbf{k}-\mathbf{k}')$ does not vanish in the vicinity of $q=0$, we can show graphically that the equation

$$1-\lambda_2(\mathbf{k}')\Pi_{+-}(\Omega)\sum_{\mathbf{q}}F^0(\mathbf{q},\Omega)\lambda_2(\mathbf{q})\Delta(\mathbf{k}-\mathbf{q})=0$$

does not have the solution $\Omega=0$ at any temperature. Here, $F^0(q, \Omega)=-1/(\Omega-vq)$, $\lambda\propto k^{1/2}$, and the function $\Pi_{+-}^{-1}(\Omega)$ is such that it does not vanish at $\Omega=0$. Thus, the initial assumption of the occurrence of a phase transition in paramagnetic crystals in which the distribution of paramagnetic ions does not satisfy Eq. (14) is incorrect.

It is known[14] that interaction of spins with phonons results in an effective dipole–dipole interaction between spins. Therefore, a phase transition in a paramagnetic crystal of the kind discussed here is due to the same interactions (more exactly, due to the quadrupole–quadrupole interaction). It should be noted that allowance for the intrinsic dipole–dipole interactions improves the conditions for a phase transition.[12]

We shall conclude by considering the possibility of a phase transition in the $S=1/2$ case. If we rewrite the interaction (1) in the second quantization representation, we find that the spin–phonon coupling constant is

$$\lambda'=(G'H_0/v)(\hbar\Omega p/m)^{1/2}.$$

The threshold condition (20) for $S=1/2$ becomes

$$H_0>g\mu_B mv^2/pG'^2. \quad (32)$$

Typical values of G' are of the order of 10^{-18} erg/Oe.[15] If we assume that the velocity of sound in a crystal is 5×10^5 cm/sec, the mass of a unit cell is 10^{-22} g, and $p=1/10$, we find that the threshold condition (32) becomes unrealistic: $H_0>10^7$ Oe.

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[1]A. F. Sadreev and A. P. Slivinskiĭ, Fiz. Tverd. Tela (Leningrad) **17**, 2443 (1975); **18**, 2169 (1976) [Sov. Phys. Solid State **17**, 1620 (1975); **18**, 1264 (1976)].
[2]S. A. Al'tshuler and B. M. Kozyrev, Elektronnyĭ paramagnitnyĭ rezonans (Electron Paramagnetic Resonance), 2nd ed., Nauka, M., 1972.
[3]L. D. Landau and E. N. Lifshitz, Statisticheskaya fizika, Nauka, M., 1964 (Statistical Physics, 2nd ed., Pergamon Press, Oxford, 1969).
[4]N. G. Koloskova, Fiz. Tverd. Tela (Leningrad) **5**, 61 (1963) [Sov. Phys. Solid State **5**, 40 (1963)].
[5]J. W. Tucker and V. W. Rampton, Microwave Ultrasonics in Solid State Physics, North-Holland, Amsterdam, 1972 (Russ. Transl., Mir, M., 1975).
[6]A. Z. Patashinskiĭ and V. L. Pokrovskiĭ, Fluktuatsionnaya teoriya fazovykh perekhodov (Fluctuation Theory of Phase Transitions), Nauka, M., 1975.
[7]A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinskiĭ, Metody kvantovoĭ teorii polya v statisticheskoĭ fizike, Izd. AN SSSR, M., 1962 (Methods of Quantum Field Theory in Statistical Physics, Prentice-Hall, Englewood Cliffs, N. J., 1963).
[8]H. Keiter, Z. Phys. **213**, 466 (1968).
[9]B. J. McKenzie and G. E. Stedman, J. Phys. A **9**, 187 (1976).
[10]A. F. Sadreev and S. N. Martynov, Fiz. Tverd. Tela (Leningrad) **19**, 744 (1977) [Sov. Phys. Solid State **19**, 431 (1977)].
[11]M. M. Zaitov, Zh. Eksp. Teor. Fiz. **56**, 88 (1969) [Sov. Phys. JETP **29**, 49 (1969)].
[12]Yu. A. Kudenko, A. F. Sadreev, and A. P. Slivinskiĭ, Preprint IFSO-39F, 1976.
[13]N. N. Bogolyubov, Sobranie sochineniĭ (Collected Works), Vol. 3, Naukova Dumka, Kiev, 1971.
[14]L. K. Aminov and B. I. Kochelaev, Zh. Eksp. Teor. Fiz. **42**, 1303 (1962) [Sov. Phys. JETP **15**, 903 (1962)].
[15]S. A. Al'tshuler, B. I. Kochelaev, and A. M. Leushin, Usp. Fiz. Nauk **75**, 459 (1961) [Sov. Phys. Usp. **4**, 880 (1962)].

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