

# THE INFLUENCE OF THE SPIN-PHONON INTERACTION IN A FERROMAGNET ON THE CURIE TEMPERATURE

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In the first order perturbation theory with respect to the Hamiltonian of the spin-phonon interaction  $H_2$  and in the second order with respect to  $H_1$  the corresponding shift of the Curie temperature  $T_c^0 \rightarrow T_c$  has been found;  $H_n$  contains the  $n$ -th power of thermal displacement. The first order correction to  $T_c^0$  increases it, while the second order one decreases  $T_c^0$ ; the total effect of the spin-phonon interaction makes the Curie temperature higher.

## 1. Introduction

The interaction of the spin system of a ferromagnet with other, non-magnetic degrees of freedom of crystal affects the relaxational and thermodynamical characteristics of the system. The corresponding effects in the case of relaxational characteristics have been considered in numerous papers. While the influence of these interactions on the thermodynamical characteristics of the spin system (the static spin correlators, the Curie temperature  $T_c^0$ ) has not been actually considered. As to the spin-phonon interaction its effect on thermodynamics of the spin system of the Heisenberg ferromagnet has been considered in [1], [2].

Szaniecki [1] showed the effect of the spin-phonon interaction on the spontaneous magnetization of the Heisenberg model. In the same model Kumar [2] studied the renormalization  $T_c^0 \rightarrow T_c$  of the Curie temperature (CT) due to the spin-phonon interaction. It appears that if one has in mind the vicinity of the Curie point a systematic study of the influence of the spin-phonon interaction on the thermodynamics of the spin system in this temperature range should be naturally started with considering the corresponding renormalization of  $T_c^0$ . This is the aim of present work. However, before we formulate the problem in more precise terms we shall consider the way it has been solved in Kumar's paper [2]. To do this we shall introduce some notations which will be used in what follows.

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The Hamiltonian of a system of exchangeably coupled spins interacting with phonons will be written as

$$H = H_0 + H_{\text{int}}, \quad H_0 = H_p + H_s, \quad H_{\text{int}} = H_1 + H_2 \quad (1)$$

$$H_p = \sum_{kj} \hbar \omega_{kj} a_{kj}^+ a_{kj}, \quad H_s = - \sum_{n \neq m} J(\mathbf{R}_{nm}) S_n^\alpha S_m^\alpha \quad (2)$$

$$H_1 = - \sum_{n \neq m} \nabla^\alpha J(\mathbf{R}_{nm}) (u_n^\alpha - u_m^\alpha) S_n^\beta S_m^\beta,$$

$$H_2 = - \frac{1}{2} \sum_{n \neq m} \nabla^\alpha \nabla^\beta J(\mathbf{R}_{nm}) (u_n^\alpha - u_m^\alpha) (u_n^\beta - u_m^\beta) S_n^\gamma S_m^\gamma. \quad (3)$$

Here  $H_p$  is the lattice Hamiltonian in harmonic approximation, so that  $a_{kj}$  is the phonon destruction operator of the  $j$ -th mode with a wave vector  $\mathbf{k}$  and the frequency  $\omega_{kj}$ ;  $H_s$  is the Heisenberg exchange Hamiltonian, so that  $J(\mathbf{R}_{nm}) \equiv J(\mathbf{R}_n - \mathbf{R}_m)$  is the exchange integral between the  $n$ -th and  $m$ -th spins.

The terms  $H_{1,2}$  of the Hamiltonian of the spin-phonon interaction obtain, as usual, by expanding the exchange integral in a power series of thermal displacements  $u_n^\alpha$  of nuclei. In (2), (3) and further throughout the repeated Greek indices of the Cartesian coordinates mean summation.

In Kumar's paper [2] in the formalism of two-time retarded and advanced Green functions [3] the mass operator for the Greenian<sup>1</sup>  $G_q(\omega) = \langle\langle S_q^+ | S_q^- \rangle\rangle_\omega$ ,  $S_q^\alpha = N^{-1/2} \sum_n \exp(-i\mathbf{q} \cdot \mathbf{R}_n) S_n^\alpha$ ,  $S_q^\pm = S_q^x \pm iS_q^y$  is obtained in the second order perturbation theory with respect to the Hamiltonian<sup>2</sup>  $H_1$ . However, he does not take into account the spin-phonon contributions to the Greenian  $g_q(\omega) = \langle\langle [S_q^+, H_s] | S_q^- \rangle\rangle_\omega$ . The Green functions  $g_q(\omega)$  and  $\langle\langle [[S_q^+, H_1], H_1] | S_q^- \rangle\rangle_\omega$  were decoupled in [2] in a different way. Now, the static spin correlator  $Q(\mathbf{q}) = \langle S_q^Z S_{-q}^Z \rangle = \text{Tr}[S_q^Z S_{-q}^Z \exp(-\beta H)] [\text{Tr} \exp(-\beta H)]^{-1}$ ,  $\beta = (k_B T)^{-1}$  was decoupled in [2] into the products of  $\langle S_q^Z \rangle = \sqrt{N} \Delta(\mathbf{q}) \sigma$ ,  $\sigma = \langle S_n^Z \rangle^3$ . As a result at  $T \rightarrow T_c - 0$  when  $\sigma \rightarrow 0$  the mass operator obtained in [2] turned out to be  $\sim \sigma^2$ , the conformity principle for the results [2] proved to be violated: at  $T \leftarrow T_c$  the Greenian  $G_q(\omega)$  calculated in [2] does not pass into the one-magnon Green function<sup>4</sup> obtained in [5] in the same (second) order perturbation theory with respect to  $H_1$ . Finally, paper [2] gives only a general expression for  $T_c - T_c^0$ ; the numerical estimates of the amount of renormalization of CT being not made in [2].

In the present work we shall obtain the renormalization of CT in a perfect one-domain cubic ferroelectric due to the spin-phonon interaction (the second order perturbation theory with respect to  $H_1$  and the first with respect to  $H_2$ ). The following assumptions will be made. (a) The first random phase approximation (RPA-I) will be used so that calculating  $G_q(\omega)$  in the Green functions containing more than one spin operator on the

<sup>1</sup> The two-time Greenians are determined similarly to [3].

<sup>2</sup> The term  $H_2$  is not taken into account in [2].

<sup>3</sup> A similar procedure was used in [4] when considering  $G_q(\omega)$  taking into account the spin-phonon interaction.

<sup>4</sup> This remark also applies to the results of paper [4].

left of | the  $\langle S_k^z \rangle$  will be taken outside the Greenian sign. As in the case of  $H_{\text{int}} \equiv 0$  we have a right to expect in the RPA-I that [6]

$$G_q(\omega) = \frac{\sigma}{\pi} \frac{1}{\omega - \Omega_q}, \quad \sigma \rightarrow 0, \quad \lim_{\sigma \rightarrow 0} \frac{\Omega_q}{\sigma} \neq 0, \infty, \quad (4)$$

only now

$$\Omega_q \neq \omega_q = \frac{2\sigma}{\hbar} [J(0) - J(q)], \quad J(q) = \sum_n J(\mathbf{R}_n) e^{-iq\mathbf{R}_n}, \quad \sum_q J(q) = 0. \quad (5)$$

(b) The back influence of the spin system on phonons will be neglected (*cf.* [7]). In other words, the spin-phonon interaction is treated adiabatically, the spin system of a magnetic playing the role of a "slow" subsystem. Therefore it can be expected that the results obtained will be valid with the condition

$$T_c^0 \ll T_D, \quad (6)$$

where  $T_D$  is the Debye temperature:  $k_B T_D = \hbar \max \omega_{kj} \equiv \hbar \omega_{mj}$ . A fairly wide class of ferromagnets exists whose parameters satisfy condition (6) [8]. (c) At  $H_{\text{int}} \equiv 0$  the pair spin correlators are isotropic in  $T_c^0$ :  $\langle S_q^+ S_{-q}^- \rangle_0 = 2Q_0(q)$ , where  $\langle \dots \rangle_0$  means averaging over Gibbs' ensemble with the Hamiltonian  $H_0$ .

## 2. The second order perturbation theory

First, in the second order perturbation theory we shall take into account the interaction described by the Hamiltonian  $H_1$  (3). We shall make the canonical transformation, so that the terms linear in phonon operators should be omitted from  $H_0 + H_1$  (*cf.* [7]):

$$a_{kj} \rightarrow b_{kj} + F_{kj}. \quad (7)$$

Here the operator  $F_{kj}$  contains only spin operators. Then, as is easily seen from (1)–(3)

$$F_{kj} = \frac{1}{\sqrt{2MN\hbar\omega_{kj}^3}} \sum_{n \neq m} [v_{kj}^\alpha \nabla^\alpha J(\mathbf{R}_{nm})]^* (e^{-ik\mathbf{R}_n} - e^{-ik\mathbf{R}_m}) S_n^\beta S_m^\beta. \quad (8)$$

Here  $M$  is the mass of a unit cell. In the case of the Bravais lattices considered below the phonon polarization vectors  $v_{kj}^\alpha = -v_{-kj}^\alpha$  can be regarded as real [9]. As a result

$$\begin{aligned} H &\rightarrow H_0, \quad H_0 = H_p + H_s', \quad H_p = \sum_{kj} \hbar \omega_{kj} b_{kj}^+ b_{kj}, \\ H_s' - H_s &= -\frac{1}{2MN} \sum_{kj} \omega_{kj}^{-2} \sum_{n \neq m} \sum_{p \neq r} [v_{kj}^\alpha \nabla^\alpha J(\mathbf{R}_{nm})]^* [v_{kj}^\beta \nabla^\beta J(\mathbf{R}_{pr})] \times \\ &\quad \times (e^{-ik\mathbf{R}_n} - e^{-ik\mathbf{R}_m}) (e^{ik\mathbf{R}_p} - e^{ik\mathbf{R}_r}) S_n^\lambda S_m^\lambda S_p^\sigma S_r^\sigma = \\ &= -\frac{1}{N} \sum_{k_1 k_2 k_3 k_4} V(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4) \Delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 + \mathbf{k}_4) S_{k_1}^\lambda S_{k_2}^\lambda S_{k_3}^\sigma S_{k_4}^\sigma \end{aligned} \quad (9)$$

$$\begin{aligned}
V(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4)\Delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 + \mathbf{k}_4) = & -\frac{1}{2M} \sum_{kj} \frac{1}{\omega_{kj}^2} [A_{kj}(\mathbf{k}_1) + \\
& + A_{kj}(\mathbf{k}_2)] [A_{kj}(\mathbf{k}_3) + A_{kj}(\mathbf{k}_4)] \Delta(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}) \Delta(\mathbf{k}_3 + \mathbf{k}_4 + \mathbf{k}), \\
A_{kj}(\boldsymbol{\kappa}) = & (\boldsymbol{\kappa} v_{kj}) J(\boldsymbol{\kappa}).
\end{aligned} \tag{10}$$

With the assumptions (a), (b), (c) at  $T = T_c$  we obtain from (9), (10)

$$\begin{aligned}
\frac{\Omega_q - \omega_q}{\sigma} = & \frac{4}{M\hbar N} \sum_{kj} \frac{1}{\omega_{kj}^2} [A_{kj}(\mathbf{k}) - A_{kj}(\mathbf{q}) - A_{kj}(\mathbf{k} - \mathbf{q})] \{Q(\mathbf{k})A_{kj}(\mathbf{k}) + \\
& + Q(\mathbf{k} - \mathbf{q}) [A_{kj}(\mathbf{q}) + A_{kj}(\mathbf{k} - \mathbf{q})]\},
\end{aligned} \tag{11}$$

$Q(\mathbf{q}) = \lim_{\sigma \rightarrow 0} [\sigma(\beta_c \hbar \Omega_q)^{-1}] \rightarrow Q_0(\mathbf{q}) = (2\beta_c^0)^{-1} [J(0) - J(\mathbf{q})]^{-1}$  according to (4). This substitution is necessary in (11) since we limit ourselves to the second order perturbation theory with respect to  $H_1$ . At  $a\mathbf{q} \ll 1$  ( $a$  is the lattice constant) in cubic crystals where  $\hbar\omega_q = 2\sigma Ja^2q^2$ ,  $J$  is the exchange integral,  $Z$  is the coordination number. Similarly,  $\hbar\Omega_q = 2\sigma Ia^2q^2$ ,  $I = J(1 - \Delta)$ . By this the relative renormalization of CT  $(T_c - T_c^0)(T_c^0)^{-1} = -\Delta$  is related to the renormalization of the exchange integral to which the CT is proportional. According to the definition of  $\Delta$  we obtain from (11):

$$\begin{aligned}
\Delta = & \frac{1}{3MNa^2\beta_c^0 J} \sum_{kj} \frac{1}{\omega_{kj}^2 [J(0) - J(\mathbf{k})]} \left\{ A_{kj}(\mathbf{k}) (\nabla^\alpha)^2 A_{kj}(\mathbf{k}) + \right. \\
& \left. + [\nabla^\alpha A_{kj}(\mathbf{k}) - \nabla^\alpha A_{kj}(\mathbf{k})|_{k=0}] \left[ \nabla^\alpha A_{kj}(\mathbf{k}) - \nabla^\alpha A_{kj}(\mathbf{k})|_{k=0} + \frac{A_{kj}(\mathbf{k}) \nabla^\alpha J(\mathbf{k})}{J(0) - J(\mathbf{k})} \right] \right\}
\end{aligned} \tag{12}$$

$v_{kj}^\alpha$  in  $A_{kj}(\mathbf{k})$  must not be differentiated with respect to  $\mathbf{k}$  in this formula. For a numerical estimate of the quantity we use in (12) the spherical approximation for  $J(\mathbf{k})$  and the isotropic Debye model for the phonon spectrum of a crystal when  $\omega_{kj} = ck$ ,  $v_{kj}^\alpha = k^\alpha k^{-1}$ ; for transversal phonons  $k_\alpha v_{kj}^\alpha = 0$ . Then

$$\Delta = \frac{T_c^0}{T_D} \eta f_1(Z), \quad f_1(Z) = \frac{1}{3\pi^2} \frac{v}{a^3} (11\pi^2 - 2Z \sqrt[3]{6\pi^2}), \quad \eta = \frac{\hbar k_m}{Mc}, \tag{13}$$

where  $k_m = \omega_{mj}c^{-1} = \max k$ ,  $a^3 k_m^3 = 6\pi^2$ ,  $v$  is the volume of a unit cell. For three types of cubic lattices, according to (13),  $f_1(6) = 2.16$ ;  $f_1(8) = 0.83$ ;  $f_1(12) = 0.17$ . If evaluating the integral in (12) we abandon the approximation of small  $ak$ , isotropy and the nearest neighbour approximation  $|f_1(Z)| = O(1)$  for  $\Delta$  in the formula (13). Since in the given order perturbation theory, by definition,  $|\Delta| \ll 1$  condition (6) follows from (13) as it must. Now, we shall note that  $\Delta > 0$ , *i. e.* when the spin-phonon interaction  $H_1$  (3) is "turned on" the CT decreases. In this connection we shall note that the effective Hamiltonian  $H'_s$  (9) resembles the biquadratic exchange Hamiltonian. As shown in [10] in the constant coupling approximation [11] the CT decreases with increasing the integral of this exchange  $J'$  (having the same sign as  $J$ ). Further, as is shown in [12] basing on the

static scaling theory [13] the CT linearly decreases with increasing  $J'$  in a first approximation with respect to  $J'J^{-1}$ . Thus our result (13) is in agreement with the results of investigation of the effect of biquadratic exchange on the CT.

### 3. The first order perturbation theory

Now, let  $H_{\text{int}} \rightarrow H_2$ . In the approximations (a), (b), (c) we shall obtain the expression (4) where  $\Omega_q \rightarrow \Omega'_q$  and

$$\frac{\Omega'_q - \omega_q}{\sigma} = -\frac{1}{MN} \sum_{kj} \frac{1}{\omega_{kj}} \text{cth} \frac{\beta_c^0 \hbar \omega_{kj}}{2} [(k^\alpha v_{kj}^\alpha + q^\alpha v_{kj}^\alpha)^2 J(\mathbf{k} + \mathbf{q}) - (q^\alpha v_{kj}^\alpha)^2 J(\mathbf{q}) - (k^\alpha v_{kj}^\alpha)^2 J(\mathbf{k})]. \quad (14)$$

Hence we find the appropriate renormalization of the CT for cubic crystals:

$$\Delta' = \frac{\hbar}{6MNa^3J} \sum_{kj} \frac{1}{\omega_{kj}} \text{cth} \frac{\beta_c^0 \hbar \omega_{kj}}{2} [2k^\beta v_{kj}^\beta \nabla^\alpha J(\mathbf{k}) + \frac{1}{2}(k^\beta v_{kj}^\beta)^2 (\nabla^\alpha)^2 J(\mathbf{k}) + J(\mathbf{k}) - J(0)]. \quad (15)$$

Clearly, this expression can be written as

$$\Delta' = \eta f_2(Z), \quad |f_2(Z)| = O(1). \quad (16)$$

Let us assume for a moment that  $T_c^0 \gg T_D$ . Then, according to the (13), (16)  $|\Delta| \gg |\Delta'|$  must be. On the other hand, in this case according to the definition of  $T_D$  we can make a substitution  $\text{cth} (\frac{1}{2}\beta_c^0 \hbar \omega_{kj}) \rightarrow 2(\beta_c^0 \hbar \omega_{kj})^{-1}$  in (14), (15), after which it is seen that  $|\Delta| \sim |\Delta'|$  from comparison (12), (15) and we come to a contradiction. Consequently, results (12), (15) "automatically" include the condition of adiabaticity (6) for the renormalization of CT. With this condition as follows from (15) in the isotropic Debye model

$$f_2(Z) = -5 \frac{v}{a^3} \left[ \frac{1}{4} + \frac{2\pi^4}{15} \left( \frac{T_c^0}{T_D} \right)^4 \right] \rightarrow -\frac{5}{4} \frac{v}{a^3}, \quad (17)$$

so that  $f_2(6) = -1.25$ ;  $f_2(8) = -0.625$ ;  $f_2(12) = -0.313$ . It is seen from (13), (16) that  $|\Delta| \ll |\Delta'|$ . Consequently, under condition (6) the spin-phonon interaction leads to increasing CT as compared with  $T_c^0$ .

For various values of the crystal parameters  $\eta \sim 10^{-2} - 10^{-3}$ . Thus, the renormalization of the CT owing to the spin-phonon interaction accounts for several percent at best. With the condition (6) it does not depend on the atomic spin  $S$  magnitude and the exchange integral  $J$ .

For crystals whose parameters obey the condition (6) it is difficult to single  $T_c^0$  out of the experimentally measured value of  $T_c$ . Therefore it should be taken into consideration that if the CT is interpreted as characteristic of a "purely" spin system there is an *a priori* error of the order of  $\pm \Delta'$  in the experimental value of the CT.

It follows from comparison of results (13), (16) that the parameter  $\eta$  plays the role of the effective coupling constant when considering the influence of the spin-phonon interaction on the spin thermodynamic characteristics of a magnetic (*cf.* [14]). It means that in the same order (*e. g.* the second order) perturbation theory the corresponding "spin-phonon" correction from the contribution of  $H_n$  to  $H_{\text{int}}$  if at all non-zero, will be proportional to  $\eta^n$ ,  $n = 1, 2, 3 \dots$

Discarding condition (6) the influence of the spin-phonon interaction on the CT can be taken into account, in principle, by means of the self-consistent Konwent and Plakida formalism [15].

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