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Path integral approach to the compressible Ising model

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Abstract. The path integral method is used to decouple the spin-phonon variables in the Wagner-Swift Hamiltonian for a compressible Ising model where the exchange interaction is expanded up to terms quadratic in displacements. A brief review is given of the earlier work. The approximations made by the previous workers to obtain the effect of the spin-phonon coupling on the Debye-Waller factor and the phonon frequencies are shown to correspond to the lowest-order terms in the cumulant expansion in the present approach. Finally the procedure to extend the formalism to anharmonic crystals is outlined.

Introduction

There is clear experimental evidence that the magnetic substances show an anomalous volume expansion in the neighbourhood of the transition temperature (Gonano *et al* 1968, Donaldson and Lanchester 1968). This volume expansion can be understood on the basis of the coupling between a system of interacting ions and interacting spins. The coupling is mainly due to the fact that the exchange interaction between the spins located on two ions is a fairly strong function of the inter-ionic distance. The distance is modulated by the lattice vibrations which, therefore, couple to the spins. The effect of the spin-phonon coupling on the magnetic properties of crystals on the basis of an Ising model has been the subject of considerable interest in recent years (Matsudaira 1968, Bolton and Lee 1970, Lee and Bolton 1971, Horner 1972, Salinas 1973, Aharony 1973). Spin-phonon coupling has also been found to effect the Mössbauer spectrum (Bashkirov and Selyutin 1968, Nandwani and Puri 1972) and the elastic constants (Wagner and Swift 1970).

The traditional approach to study the influence of spin-phonon coupling is to expand the exchange and the lattice energies in terms of the displacements of the ions around the equilibrium positions. The coupling between the displacements and the spins is precisely what makes the problem difficult to solve. However, if we approximate the Hamiltonian of the system by retaining only the quadratic term in the potential energy and the linear term in the exchange energy, it is possible to completely decouple the spin and the phonon variables and obtain an effective Hamiltonian. This has been shown previously by Matsudaira (1968), Wagner and Swift (1970) and Bolton and Lee (1970). To this order of approximation the Ising exchange interaction does not lead to a renormalization of phonon frequencies (Oitmaa 1974). However, one expects that the phonon frequencies should be sensitive to the ordering of the spins. To obtain this

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effect in the Ising model it is necessary, therefore, to expand the exchange energy to second order in ionic displacements. Such a term was in fact considered in an approximate way by Bashkirov and Selyutin (1968) as contributing to the Debye–Waller factor, and by Wagner and Swift (1970) as contributing to the elastic constants. This term was shown by Wagner and Swift to be important also in preserving the symmetry properties of the elastic constants. To our knowledge no effort has so far been made to obtain an effective Hamiltonian in the presence of the quadratic term in the exchange interaction.

The purpose of this paper is to derive an effective spin and an effective lattice Hamiltonian by decoupling the spin and the phonon variables of an Ising model in which the exchange interaction is expanded up to second order in displacements. To achieve this, we use the path integral method first introduced by Feynman (1948). One of the advantages of this method is that the operators are turned into classical variables. Since the original formulation, this method has been applied successfully to several problems such as liquid helium (Feynman 1953), polarons (Feynman 1955) and anharmonic crystals (Samathiyakanit and Glyde 1973).

The paper can also be regarded as an application of the path integral method to a new area. To obtain the effective Hamiltonian we first eliminate the linear terms by averaging over the quadratic terms. Taking advantage of the Gaussian averaging, a cumulant expansion of the average is made. In the second step we average the displacement term over that part of the Hamiltonian which contains only the spins. This immediately decouples the partition function. The present problem is in a way similar to the polaron problem studied by Feynman (1955). The important advantage of this kind of approach is that successive orders of approximation can be employed to improve upon the results. It is shown here that the previous work on the Debye–Waller factor by Bashkirov and Selyutin (1968) and on the shift of the phonon frequencies by Oitmaa (1974) corresponds to retaining the lowest-order cumulants in the present treatment. We also discuss the possible extension of the method to include the anharmonic effects.

2. Wagner–Swift Hamiltonian

We consider a crystal of N identical atoms. For simplicity we assume one atom per unit cell. The formalism can easily be extended to non-primitive lattices. If we expand the potential energy Φ and the exchange energy Ψ in powers of displacements, the Hamiltonian of the lattice in the harmonic approximation may be written as

$$H = (1/2M) p_\alpha(l)p_\alpha(l) + \Phi_0 + \Phi_\alpha(l)u_\alpha(l) + \frac{1}{2}\Phi_{\alpha\beta}(l, l')u_\alpha(l)u_\beta(l') + \Psi_0 + \Psi_\alpha(l)u_\alpha(l) + \frac{1}{2}\Psi_{\alpha\beta}(l, l') \times u_\alpha(l)u_\beta(l') - K_\alpha(l)r_\alpha(l), \quad (1)$$

where $u_\alpha(l)$ is the α Cartesian component of the displacement with respect to the equilibrium position of the l th ion, $p_\alpha(l)$ is the conjugate momentum, M is the atomic mass, Φ_0 is the static potential energy, and

$$\Psi = \frac{1}{2}J(r(i) - r(j))\sigma_i\sigma_j \quad (2)$$

is the magnetic exchange Hamiltonian in which σ_i is the single-component spin variable of the i th ion. The external force on the ion at $r(l)$ is denoted by $\mathbf{K}(l)$. A summation over repeated indices is applied everywhere. $\Phi_\alpha(l)$, $\Phi_{\alpha\beta}(l, l')$, $\Psi_\alpha(l)$ and $\Psi_{\alpha\beta}(l, l')$ are the coefficients of expansion evaluated at the equilibrium positions. $\Psi_{\alpha\beta}(l, l')$, for example,

is defined as

$$\Psi_{\alpha\beta}(l, l') = \partial^2 \Psi / \partial u_\alpha(l) \partial u_\beta(l')|_0. \quad (3)$$

In the Feynman formulation, the partition function is given by

$$Z = \text{Tr} \int d^N \mathbf{u}(i) \int \mathcal{D}^N(\mathbf{u}(i, \tau)) \exp\left(-\int_0^\beta H(\tau) d\tau\right), \quad (4)$$

where $\mathcal{D}^N(\mathbf{u}(l, \tau))$ denotes the integration over all possible paths. $H(\tau)$ is obtained from H by introducing time dependence in $\mathbf{r}(l)$, $\mathbf{u}(l)$ and $\mathbf{p}(l)$ explicitly. The coefficients of expansion are assumed to be independent of time. The upper limit β of the integral means $(1/k_B T)$ and is not to be confused with the subscript β which is the cartesian component. Tr means the trace with respect to the spins. It is convenient to define

$$H_h = (1/2M)p_\alpha(l)p_\alpha(l) + \Phi_0 + \frac{1}{2}(\Phi_{\alpha\beta}(l, l') + \Psi_{\alpha\beta}(l, l'))u_\alpha(l)u_\beta(l'). \quad (5)$$

Taking advantage of the quadratic nature of H_h , the partition function may be written as

$$Z = \text{Tr} \int d^N \mathbf{u}(i) \int \mathcal{D}^N(\mathbf{u}(i, \tau)) \exp\left(-\int_0^\beta [H_h(\tau) + \Psi_0] d\tau\right) \times \left\langle \exp\left\{-\int_0^\beta d\tau[(\Phi_\alpha(l) + \Psi_\alpha(l))u_\alpha(l, \tau) - K_\alpha(l)r_\alpha(l, \tau)]\right\} \right\rangle_{H_h}, \quad (6)$$

where

$$\langle O \rangle_{H_h} = Z_h^{-1} \int d^N \mathbf{u}(i) \int \mathcal{D}^N(\mathbf{u}(i, \tau)) \exp\left(-\int_0^\beta H_h(\tau) d\tau\right) O, \quad (7)$$

and

$$Z_h = \int d^N \mathbf{u}(i) \int \mathcal{D}^N(\mathbf{u}(i, \tau)) \exp\left(-\int_0^\beta H_h(\tau) d\tau\right). \quad (8)$$

The interesting thing about writing the partition function in the form of equation (6) is that, on expanding the average in cumulants, only the second cumulant is non-zero. This is due to the fact that while H_h is quadratic in displacements, the exponent inside the angular brackets is linear. Equation (6), therefore, may be rewritten as

$$Z = \text{Tr} \int d^N \mathbf{u}(i) \int \mathcal{D}^N(\mathbf{u}(i, \tau)) \exp\left(-\int_0^\beta (H_h(\tau) + \Psi_0) d\tau + \frac{1}{2}\beta D_{\alpha\beta}(l, l') \times (\Phi_\alpha(l) + \Psi_\alpha(l) - K_\alpha(l))(\Phi_\beta(l') + \Psi_\beta(l') - K_\beta(l')) + \beta K_\alpha(l)R_\alpha(l)\right), \quad (9)$$

where $\mathbf{R}(l)$ is the equilibrium position of the l th ion and

$$D_{\alpha\beta}(l, l') = \int_0^\beta \langle u_\alpha(l, \tau)u_\beta(l', 0) \rangle_{H_h} d\tau. \quad (10)$$

It may be pointed out that the partition function given by equation (9) is exact for the Wagner–Swift Hamiltonian. The linear terms do not appear explicitly now. The phonon and the spin variables are still not decoupled due to the presence of $\Psi_{\alpha\beta}(l, l')$ in H_h . However, if we put $\Psi_{\alpha\beta}(l, l') = 0$, the effective spin Hamiltonian is immediately obtained from equation (9) and may be compared with the results obtained by Bolton and Lee (1970), who used a perturbation technique, or with the effective Hamiltonian obtained by Wagner and Swift (1970) with the help of a unitary transformation.

At this point a word about the equilibrium positions seems to be in order. The displacements $\mathbf{u}(l)$, as has already been pointed out, are with respect to the true equilibrium positions which can be obtained, in principle, by differentiating the total free energy

with respect to the external forces. The equilibrium positions evidently differ from the rest positions determined by the minimum of the potential energy, firstly because of the external forces and secondly because of the anomalous thermal expansion. The equilibrium condition may be written as:

$$R_\alpha(l) = \frac{1}{\beta} \frac{\partial \ln Z}{\partial K_\alpha(l)}, \quad (11)$$

which can be written, because of equation (9), in the form

$$\langle D_{\alpha\beta}(l, l')(\Phi_\beta(l') + \Psi_\beta(l') - K_\beta(l')) \rangle_H = 0, \quad (12)$$

where the average is with respect to the total Hamiltonian and is defined by

$$\langle O \rangle_H = Z^{-1} \text{Tr} \int d^N \mathbf{u}(i) \int \mathcal{D}^N(\mathbf{u}(i, \tau)) \exp\left(-\int_0^\beta H(\tau) d\tau\right) O. \quad (13)$$

The isothermal elastic constants can be obtained by making an infinitesimal homogeneous deformation, $R_\alpha(l) \rightarrow R_\alpha(l) + \delta R_\alpha(l)$, and by varying the external force $K_\alpha(l) \rightarrow K_\alpha(l) + \delta K_\alpha(l)$. The temperature is kept constant. The expansion coefficients $\Phi_\alpha(l)$ and $\Psi_\alpha(l)$ will change accordingly. However, $D_{\alpha\beta}(l, l')$ which depends upon the force constants $\Phi_{\alpha\beta}(l, l')$ and $\Psi_{\alpha\beta}(l, l')$ is to be kept constant in the harmonic approximation. The anharmonic terms should be included in a consistent manner if the strain dependence of the force constants is to be included. In the presence of the deformation, the equilibrium condition, therefore, becomes

$$\langle D_{\alpha\beta}(l, l')[(\Phi_{\beta\gamma}(l', l'') + \Psi_{\beta\gamma}(l', l''))\delta R_\gamma(l'') - \delta K_\beta(l'')] \rangle_{H'} = 0, \quad (14)$$

where H' is the new Hamiltonian obtained by changing $R_\alpha(l)$ and $K_\alpha(l)$ to their new values. If we retain only those terms which are linear in the deformation, the new equilibrium positions are given by

$$\begin{aligned} \delta R_\alpha(l) = & -\langle D_{\alpha\beta}(l, l') \rangle_H \delta K_\beta(l') + \beta \delta K_\alpha(m) \langle D_{\alpha\beta}(l, l')(\Phi_\beta(l') + \Psi_\beta(l') - K_\beta(l')) \\ & \times D_{\alpha'\beta'}(m, n)(\Phi_{\beta'}(n) + \Psi_{\beta'}(n) - K_{\beta'}(n)) \rangle_H. \end{aligned} \quad (15)$$

It is evident from equation (10) that $D_{\alpha\beta}(l, l')$ depends on the spin configuration. Wagner and Swift (1970) approximate it by its thermal average. There is no need of this approximation if one neglects the second-order term in the exchange interaction. As has been pointed out by them it is better to approximate $D_{\alpha\beta}(l, l')$ than to neglect the $\Psi_{\alpha\beta}(l, l')$ term. This problem has been studied in detail by Wagner and Swift (1970) and we shall not dwell on this any more. In the following section we try to decouple the spin and the phonon variables in equation (9).

3. Effective Hamiltonian

It is clear that the spin and the phonon variables are coupled in the $\Psi_{\alpha\beta}(l, l')$ term in equation (9). In order to achieve the decoupling, we rewrite the partition function in the form

$$\begin{aligned} Z = & \text{Tr} \exp(-\beta H_s) \int d^N \mathbf{u}(i) \int \mathcal{D}^N(\mathbf{u}(i, \tau)) \\ & \times \left\langle \exp\left[-\int_0^\beta H_h(\tau) d\tau\right] \right\rangle_{H_s} \exp(\beta K_\alpha(l) R_\alpha(l)), \end{aligned} \quad (16)$$

where the purely spin-dependent Hamiltonian, H_s , is defined as

$$H_s = \Psi_0 - \frac{1}{2} \mathcal{D}_{\alpha\beta}(l, l') (\Phi_\alpha(l) + \Psi_\alpha(l) - K_\alpha(l)) (\Phi_\beta(l') + \Psi_\beta(l') - K_\beta(l')), \quad (17)$$

and

$$\langle O \rangle_{H_s} = [\text{Tr} \exp(-\beta H_s) O] / \text{Tr} \exp(-\beta H_s). \quad (18)$$

The advantage of writing the partition function in the form of equation (16) is that Z can be looked upon as a product of two partition functions, one involving only the spin variables and the other involving only the displacements. We may, therefore, write equation (16) as

$$Z = Z_s Z_L, \quad (19)$$

where

$$Z_s = \text{Tr} \exp(-\beta H_s) \quad (20)$$

and

$$Z_L = \int d^N \mathbf{u}(i) \int \mathcal{D}^N(\mathbf{u}(i, \tau)) \left\langle \exp\left[-\int_0^\beta H_h(\tau) d\tau\right] \right\rangle_{H_s} \exp(\beta K_\alpha(l) R_\alpha(l)). \quad (21)$$

Expanding the average in cumulants, Z_L becomes

$$\begin{aligned} Z_L = \int d^N \mathbf{u}(i) \int \mathcal{D}^N(\mathbf{u}(i, \tau)) \exp\left[-\int_0^\beta H_{Lh}(\tau) d\tau - \frac{1}{2} \int_0^\beta u_\alpha(l, \tau) u_\beta(l', \tau) \langle \Psi_{\alpha\beta}(l, l') \rangle_{H_s} d\tau \right. \\ \left. + \frac{1}{4} \left(\int_0^\beta u_\alpha(l, \tau) u_\beta(l', \tau) d\tau \right) \left(\int_0^\beta \mu_\alpha(m, \tau) u_\beta(n, \tau) d\tau \right) \left[\langle \Psi_{\alpha\beta}(l, l') \Psi_{\alpha'\beta'}(m, n) \rangle_{H_s} \right. \right. \\ \left. \left. - \langle \Psi_{\alpha\beta}(l, l') \rangle_{H_s} \langle \Psi_{\alpha'\beta'}(m, n) \rangle_{H_s} \right] + \dots + \beta K_\alpha(l) R_\alpha(l) \right], \quad (22) \end{aligned}$$

where H_{Lh} is the phonon Hamiltonian in the harmonic approximation and is given by

$$H_{Lh} = (1/2M) p_\alpha(l) p_\alpha(l) + \Phi_0 + \frac{1}{2} \Phi_{\alpha\beta}(l, l') u_\alpha(l, \tau) u_\beta(l', \tau). \quad (23)$$

The effective phonon Hamiltonian can immediately be written down. So far our treatment has been exact. In practice, as we shall see later, one has to approximate Z_L . Depending on what particular physical property one is interested in, one can start either with Z_L or with Z_s . First we try to study the effective spin Hamiltonian H_s in some more detail.

3.1. The effective spin Hamiltonian

It may easily be verified that $D_{\alpha\beta}(l, l')$ that occurs in equation (17) depends only on the spin variables. H_s , therefore, depends only on the spin configuration. In order to obtain the explicit spin dependence of H_s , we proceed as follows.

$D_{\alpha\beta}(l, l')$ can be evaluated analytically by solving the path integral (Feynman 1948) after expressing the displacements in terms of the normal coordinates,

$$\mathbf{u}(l) = (1/NM)^{1/2} \sum_{\mathbf{q}, \lambda} Q(\mathbf{q}, \lambda) \mathbf{e}(\mathbf{q}, \lambda) \exp(i\mathbf{q} \cdot \mathbf{R}(l)), \quad (24)$$

where \mathbf{q} is the reciprocal lattice vector in the first Brillouin zone. $\mathbf{e}(\mathbf{q}, \lambda)$ is the polarization vector corresponding to the wavevector \mathbf{q} and the branch λ . For a primitive lattice,

$\lambda = 1, 2, 3$. $Q(\mathbf{q}\lambda)$ are the normal coordinates. $D_{\alpha\beta}(l, l')$ then becomes

$$D_{\alpha\beta}(l, l') = (1/NM) \sum_{\mathbf{q}, \lambda} (1/\omega^2(\mathbf{q}\lambda)) e_{\alpha}(\mathbf{q}\lambda) e_{\beta}^*(\mathbf{q}\lambda) \exp[i\mathbf{q} \cdot (\mathbf{R}(l) - \mathbf{R}(l'))] \quad (25)$$

where $\omega^2(\mathbf{q}\lambda)$ are the eigenvalues of the dynamical matrix

$$M_{\alpha\beta}(\mathbf{q}) = (1/M) \{ \Phi_{\alpha\beta}(l, l') + \Psi_{\alpha\beta}(l, l') \} \exp[i\mathbf{q} \cdot (\mathbf{R}(l') - \mathbf{R}(l))]. \quad (26)$$

It is convenient to write ω^2 as

$$\omega^2(\mathbf{q}\lambda) = \omega_{\perp}^2(\mathbf{q}\lambda) + \omega_s^2(\mathbf{q}\lambda), \quad (27)$$

where $\omega_{\perp}^2(\mathbf{q}\lambda)$ is the eigenvalue of the dynamical matrix without the exchange interaction and $\omega_s^2(\mathbf{q}\lambda)$ is the increment in the eigenvalue due to spin-phonon coupling. The magnitude of ω_s^2 is likely to be small compared with ω_{\perp}^2 (Nandwani and Puri 1972, Oitmaa, 1974). Therefore, it seems reasonable to write $(1/\omega^2)$ in equation (25) as:

$$\{ \omega(\mathbf{q}\lambda) \}^{-2} \simeq \{ \omega_{\perp}(\mathbf{q}\lambda) \}^{-2} [1 - (\omega_s(\mathbf{q}\lambda)/\omega_{\perp}(\mathbf{q}\lambda))^2 + (\omega_s(\mathbf{q}\lambda)/\omega_{\perp}(\mathbf{q}\lambda))^4]. \quad (28)$$

From equations (17), (25) and (28) we obtain by straightforward algebra

$$H_s = \Psi_0 - \sum_{\mathbf{q}, \lambda} (2NM\omega_{\perp}^2(\mathbf{q}\lambda))^{-1} (\Phi_{\alpha}(l) + \Psi_{\alpha}(l) - K_{\alpha}(l)) (\Phi_{\beta}(l') + \Psi_{\beta}(l') - K_{\beta}(l')) e_{\alpha}(\mathbf{q}\lambda) e_{\beta}^*(\mathbf{q}\lambda) \\ \times [1 - (\omega_s(\mathbf{q}\lambda)/\omega_{\perp}(\mathbf{q}\lambda))^2 + (\omega_s(\mathbf{q}\lambda)/\omega_{\perp}(\mathbf{q}\lambda))^4] \exp[i\mathbf{q} \cdot (\mathbf{R}(l) - \mathbf{R}(l'))]. \quad (29)$$

In order to depict the dependence of H_s explicitly on the spin configuration, we write with the help of equations (2), (26) and (27),

$$\Psi_{\alpha\beta}(l, l') = \frac{1}{2} J_{\alpha\beta}^{ij}(l, l') \sigma_i \sigma_j, \quad (30)$$

and

$$\omega_s^2(\mathbf{q}\lambda) = T_{ij}(\mathbf{q}\lambda) \sigma_i \sigma_j, \quad (31)$$

where $J_{\alpha\beta}^{ij}(l, l')$ is defined in a similar manner as $\Psi_{\alpha\beta}(l, l')$ and

$$T_{ij}(\mathbf{q}\lambda) = (1/2M) e_{\alpha}^*(\mathbf{q}\lambda) J_{\alpha\beta}^{ij}(l, l') e_{\beta}(\mathbf{q}\lambda) \exp[i\mathbf{q} \cdot (\mathbf{R}(l') - \mathbf{R}(l))]. \quad (32)$$

Re-arranging the terms in equation (29) according to the number of spin operators appearing, we obtain

$$H_s = H_0 + H_2 + H_4 + \dots, \quad (33)$$

where

$$H_0 = - \sum_{\mathbf{q}, \lambda} (2NM\omega_{\perp}^2(\mathbf{q}\lambda))^{-1} (\Phi_{\alpha}(l) - K_{\alpha}(l)) (\Phi_{\beta}(l') - K_{\beta}(l')) e_{\alpha}(\mathbf{q}\lambda) e_{\beta}^*(\mathbf{q}\lambda) \\ \times \exp[i\mathbf{q} \cdot (\mathbf{R}(l) - \mathbf{R}(l'))], \\ H_2 = \frac{1}{2} J_0^i \sigma_i \sigma_j - \sum_{\mathbf{q}, \lambda} (2NM\omega_{\perp}^2(\mathbf{q}\lambda))^{-1} e_{\alpha}(\mathbf{q}\lambda) e_{\beta}^*(\mathbf{q}\lambda) \exp[i\mathbf{q} \cdot (\mathbf{R}(l) - \mathbf{R}(l'))] \\ \times [J_{\alpha}^{ij}(l) (\Phi_{\beta}(l') - K_{\beta}(l')) - (1/\omega_{\perp}^2(\mathbf{q}\lambda)) T_{ij}(\mathbf{q}\lambda) \\ \times (\Phi_{\alpha}(l) - K_{\alpha}(l)) (\Phi_{\beta}(l') - K_{\beta}(l'))] \sigma_i \sigma_j, \\ H_4 = - \sum_{\mathbf{q}, \lambda} (2NM\omega_{\perp}^2(\mathbf{q}\lambda))^{-1} e_{\alpha}(\mathbf{q}\lambda) e_{\beta}^*(\mathbf{q}\lambda) \exp[i\mathbf{q} \cdot (\mathbf{R}(l) - \mathbf{R}(l'))] \\ \times [\frac{1}{4} J_{\alpha}^{ij}(l) J_{\beta}^{mn}(l') - (1/\omega_{\perp}^2(\mathbf{q}\lambda)) J_{\alpha}^{ij}(l) T_{ij}(\mathbf{q}\lambda) \\ \times (\Phi_{\beta}(l') - K_{\beta}(l')) + (1/\omega_{\perp}^4(\mathbf{q}\lambda)) T_{ij}(\mathbf{q}\lambda) T_{mn}(\mathbf{q}\lambda) \\ \times (\Phi_{\alpha}(l) - K_{\alpha}(l)) (\Phi_{\beta}(l') - K_{\beta}(l'))] \sigma_i \sigma_j \sigma_m \sigma_n. \quad (34)$$

In writing down the above equations, we have made use of the following property of the eigenvectors:

$$e(-q\lambda) = e^*(q\lambda). \quad (35)$$

It may be observed that H_0 is spin independent, H_2 contains two spins and H_4 contains products of four spins. In general, of course, H_s contains higher-order terms also which may easily be obtained by including higher-order terms in the expansion of $1/\omega^2(q\lambda)$ in equation (28). However, we feel that the 6-spin terms may not be very important.

To compare our results with those obtained by Bolton and Lee (1970) we simply put $T_{ij}(q\lambda) = 0$ in equations (34). This is equivalent to neglecting the second-order term in the exchange interaction. To this order of approximation H_6 and the higher-order terms in equation (33) are exactly zero. If we do this we obtain

$$H_s = H_0 + \frac{1}{2}J_0^{ij}\sigma_i\sigma_j - \sum_{q,\lambda} \{2NM\omega_L^2(q\lambda)\}^{-1} e_\alpha(q\lambda)e_\beta^*(q\lambda) \exp[-iq \cdot (R(l) - R(l'))] \\ \times [J_\alpha^{ij}(l)(\Phi_\beta(l') - K_\beta(l')) + \frac{1}{4}J_\alpha^{ij}(l)J_\beta^{mn}(l')] \sigma_i\sigma_j\sigma_m\sigma_n. \quad (36)$$

Equation (36) differs from the expression for the effective spin Hamiltonian obtained by Bolton and Lee (1970) due to the fact that they expand the potential and the exchange energy about the rest positions rather than the equilibrium positions. If we put $\Phi_\alpha(l) = K_\alpha(l)$ in the above expressions, it reduces exactly to their expression. However, we feel that the effective spin Hamiltonian obtained here is more general and consistent with the fact that the equilibrium positions depend upon the spin configuration.

3.2. Effective phonon Hamiltonian

If one is interested in the effect of spin-phonon coupling on the phonons, it is convenient to start with equation (22). If we define the effective phonon Hamiltonian H_L by

$$Z_L = \int d^N \mathbf{u}(i) \int \mathcal{D}^N(\mathbf{u}(i, \tau)) \exp\left[-\int_0^\beta H_L(\tau) d\tau\right], \quad (37)$$

we obtain from equations (22) and (30)

$$H_L = H_{Lh} - K_\alpha(l)R_\alpha(l) + \frac{1}{4}J_{\alpha\beta}^{ij}(l, l')u_\alpha(l)u_\beta(l') \langle \sigma_i\sigma_j \rangle_{H_s} + \frac{1}{16}J_{\alpha\beta}^{ij}(l, l')J_{\alpha'\beta'}^{i'j'}(m, n)u_\alpha(l)u_\beta(l') \\ \times \int_0^\beta u_{\alpha'}(m, \tau)u_{\beta'}(n, \tau) d\tau (\langle \sigma_i\sigma_j\sigma_{i'}\sigma_{j'} \rangle_{H_s} - \langle \sigma_i\sigma_j \rangle_{H_s} \langle \sigma_{i'}\sigma_{j'} \rangle_{H_s}) + \dots \quad (38)$$

It may be observed that H_L does not contain any explicit spin dependence. Though we started with the harmonic Hamiltonian, the effective phonon Hamiltonian contains terms which are quartic and of higher in displacements. The spin-phonon coupling, therefore, can be considered as giving rise to some kind of anharmonicity. The anomalous thermal expansion could be understood as partly due to this induced anharmonicity and partly due to the effect of the coupling on the equilibrium positions.

In those cases where the spin fluctuations and the strength of the exchange coupling are not large, the second and the higher-order cumulants in equation (38) may be neglected. To this order of approximation, the effective Hamiltonian may be written, after substituting for the H_{Lh} from equation (23), as

$$H_L = (1/2M)p_\alpha(l)p_\alpha(l) + \Phi_0 - K_\alpha(l)R_\alpha(l) + \frac{1}{2}\bar{\Phi}_{\alpha\beta}(l, l')u_\alpha(l)u_\beta(l'), \quad (39)$$

where the effective force constants are defined by

$$\bar{\Phi}_{\alpha\beta}(l, l') = \Phi_{\alpha\beta}(l, l') + \frac{1}{2} J_{\alpha\beta}^{ij}(l, l') \langle \sigma_i \sigma_j \rangle_{H_s}. \quad (40)$$

It is evident from the above equations that the exchange interaction modifies the force constants and, therefore, the phonon frequencies are renormalized. Recently, Oitmaa (1974) has discussed the effect of the exchange interaction on the phonon frequencies of a simple cubic lattice with nearest-neighbour interactions. If we compare equation (4) in his paper with our results, the approximations made by him seem to correspond to the neglect of the second cumulant in the effective phonon Hamiltonian obtained here.

In the light of the above comments it is expected that the Debye-Waller factor should also depend on the strength of the spin-phonon coupling. The importance of this effect has been pointed out earlier by Bashkirov and Selyutin (1968). To obtain the Debye-Waller factor they used the Green function method and employed a low-order decoupling scheme. In the present treatment we may write down the formal expression for the exponent of the Debye-Waller factor as

$$2W = 4\pi^2 g_\alpha g_\beta \langle u_\alpha(l, \tau) u_\beta(l, \tau) \rangle_{H_L}, \quad (41)$$

where \mathbf{g} is the wavevector of the gamma ray. The path integral involved in equation (41) is difficult to solve for the general case. However, if we approximate the effective phonon Hamiltonian by equation (39), where we retained only the first cumulant, the path integral can be done analytically after making the normal mode transformation. The exponent of the Debye-Waller factor is then given by

$$2W = (2\pi^2 \hbar / NM) \sum_{\mathbf{q}, \lambda} [(\mathbf{g} \cdot \mathbf{e}(\mathbf{q}\lambda))^2 / \bar{\omega}(\mathbf{q}\lambda)] \coth(\frac{1}{2}\beta \hbar \bar{\omega}(\mathbf{q}\lambda)), \quad (42)$$

where $\bar{\omega}(\mathbf{q}\lambda)$ are the normal mode frequencies determined by the effective force constants defined in equation (40). If we compare equation (42) with the results obtained by Bashkirov and Selyutin (1968), it appears that the decoupling scheme used by them is equivalent to the approximation in which only the first cumulant in the effective spin Hamiltonian is retained.

4. The self-consistent theory

The above formalism is not very suitable for anharmonic crystals. The difficulty arises mainly due to the fact that now all the cumulants in general will appear in the expansion for the average in equation (6). When the anharmonic effects may be important, it is better to follow the self-consistent procedure given recently by Samathiyakanit and Glyde (1973). We wish to extend their formalism to include the exchange interaction. Following their notation, the general Hamiltonian may be written as:

$$H(\tau) = (1/2M) p_i(\tau) p_i(\tau) + \exp(u_i(\tau) \nabla_i) (\Phi + \frac{1}{2} J^{kl} \sigma_k \sigma_l), \quad (43)$$

where the cartesian component α and the atom index l have been sub-summed in a composite index i . For simplicity we have omitted the external forces which can always be included later. ∇_i means $\partial/\partial u_i$ evaluated at the equilibrium positions. We introduce a model Hamiltonian

$$H_0(\tau) = (1/2M) p_i(\tau) p_i(\tau) + \frac{1}{2} \int_0^\beta \hat{\Phi}_{ij}(\tau, \tau') u_i(\tau) u_j(\tau') d\tau' + \frac{1}{2} J^{kl} \sigma_k \sigma_l. \quad (44)$$

The force constants $\hat{\Phi}_{ij}(\tau, \tau')$ have been taken to be explicitly time dependent to include the possibility of phonons with a finite lifetime. On introducing the model partition function

$$Z_0 = \text{Tr} \int d^N \mathbf{u}_i \int \mathcal{D}^N(\mathbf{u}_i(\tau)) \exp\left(-\int_0^\beta H_0(\tau) d\tau\right), \quad (45)$$

we may write the partition function of the system as

$$Z = Z_0 \langle \exp[-\int_0^\beta (H(\tau) - H_0(\tau)) d\tau] \rangle, \quad (46)$$

where

$$\langle O \rangle = Z_0^{-1} \text{Tr} \int d^N \mathbf{u}_i \int \mathcal{D}^N(\mathbf{u}_i(\tau)) \exp\left(-\int_0^\beta H_0(\tau) d\tau\right) O. \quad (47)$$

On expanding the average in cumulants

$$Z = Z_0 \exp\left(-\int_0^\beta \langle H(\tau) - H_0(\tau) \rangle d\tau + \frac{1}{2} \langle [\int_0^\beta (H(\tau) - H_0(\tau)) d\tau]^2 \rangle - \langle \int_0^\beta (H(\tau) - H_0(\tau)) d\tau \rangle^2 + \dots\right). \quad (48)$$

In the first-order self-consistent theory we retain only the first cumulant in the above equation and write the free energy with the help of equations (43) and (45) as

$$-\beta F = \ln Z_0 - \int_0^\beta d\tau \langle \exp(u_i \nabla_i) (\Phi + \frac{1}{2} J^{kl} \sigma_k \sigma_l) - \frac{1}{2} \int_0^\beta d\tau' \hat{\Phi}_{ij}(\tau, \tau') u_j(\tau') - \frac{1}{2} \hat{J}^{ij} \sigma_i \sigma_j \rangle. \quad (49)$$

Expanding the average in cumulants and using the fact that, for Gaussian averaging such as appears in equation (49), only the second cumulant is non-zero, we obtain

$$-\beta F = \ln Z_0 - \int_0^\beta d\tau \int_0^\beta d\tau' [\exp(\frac{1}{2} D_{ij}(\tau, \tau') \nabla_i \nabla_j) (\Phi + \frac{1}{2} J^{kl} \langle \sigma_k \sigma_l \rangle) \delta(\tau - \tau') - \frac{1}{2} D_{ij}(\tau, \tau') \hat{\Phi}_{ij}(\tau, \tau')] + \frac{1}{2} \beta \hat{J}^{ij} \langle \sigma_i \sigma_j \rangle, \quad (50)$$

where

$$D_{ij}(\tau, \tau') = \langle u_i(\tau) u_j(\tau') \rangle. \quad (51)$$

To choose the force constants $\hat{\Phi}_{ij}(\tau, \tau')$ and \hat{J}^{ij} , we use the variational principle and minimize the free energy with respect to $\hat{\Phi}_{ij}(\tau, \tau')$ and \hat{J}^{ij} to obtain

$$\hat{\Phi}_{mn}(\tau, \tau') = \frac{1}{2} \delta(\tau - \tau') \exp(\frac{1}{2} D_{ij}(\tau, \tau') \nabla_i \nabla_j) \nabla_m \nabla_n (\Phi + \frac{1}{2} J^{kl} \langle \sigma_k \sigma_l \rangle)$$

and

$$\hat{J}^{mn} = \exp(\frac{1}{2} D_{ij}(\tau, \tau) \nabla_i \nabla_j) J^{mn}. \quad (52)$$

The above equations for the force constants must be solved self-consistently in conjunction with equation (44). It may easily be seen that, if the exchange interaction is taken to be zero, equations (52) reduce to the expression obtained by Samathiyakanit and Glyde (1973).

5. Conclusion

In the present paper we have used the path integral method to decouple the spin and the phonon variables in the Hamiltonian of a compressible Ising model where the exchange interaction is expanded up to terms quadratic in displacements. To achieve this we first eliminated the linear terms by averaging over H_h which contains only the quadratic terms. When a cumulant expansion of the average is made, only the second cumulant is non-zero due to the Gaussian averaging. In the second step we average the displacement term over H_s which depends on spins only. This enables us to write the Hamiltonian as a sum of two terms, the effective spin Hamiltonian and the effective phonon Hamiltonian. The effective spin Hamiltonian is shown to reduce to that obtained earlier by Bolton and Lee (1970) and by Wagner and Swift (1970), both of whom neglected the quadratic term in the exchange interaction.

The path integral method appears to be very useful for investigating the effect of spin-phonon coupling. An important advantage of this approach is that successive orders of approximation can be employed to improve upon the results. It has been shown here that the approximations made by Bashkirov and Selyutin (1968) in obtaining the Debye-Waller factor, and by Oitmaa (1974) in studying the effect on the phonon frequencies, correspond to retaining the lowest-order cumulant in our treatment.

The spin-phonon coupling is shown to give rise to 4-spin and higher-order spin terms in the effective spin Hamiltonian. The quadratic term in the exchange interaction not only introduces the 6-spin and higher-order terms but also modifies the coefficient of the 4-spin term. Recently, Aharony (1973) has studied the critical behaviour of magnets with linear exchange coupling in the framework of renormalization group recursion relations. Since the quadratic exchange coupling changes the coefficient of the 4-spin term, the critical behaviour may also be modified.

It is also shown that the quadratic exchange coupling induces anharmonicity in the effective phonon Hamiltonian. These anharmonic terms are important for the study of thermal expansion. Finally we have outlined the procedure to treat the exchange coupling in anharmonic crystals.

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