

# ON THE LINEAR APPROXIMATION IN DYSON'S GENERALIZED SPIN WAVE FORMALISM

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(Received August 8, 1966)

The refinement of Dyson's spin wave formalism for bilinear spin Hamiltonians as proposed in a former paper [1a] is here extended to the case of fourth-order interactions. The external magnetic field is included. Without specifying the interaction tensors and the crystal lattice, and without limiting the interaction to any particular neighbourhood the conditions are derived under which the bilinear part of the ideal spin wave Hamiltonian can be diagonalized by employing Bogolyubov's transformation. Instead of using the standard or any particular spin wave representation the considerations are carried through for the wider class of representations corresponding to arbitrary inhomogeneous rotations of the lattice spins. This allows to choose in each particular case a representation that is most convenient (or otherwise preferable) in calculating the partition function (or any other thermodynamic quantity). Within the linear Bloch approximation results obtained by other authors in the standard approach are shown to be easily derivable from our formalism.

## 1. Introduction

Recently [1a], a refinement of Dyson's spin wave formalism was proposed, by introducing generalized spin wave representations generated from the standard one by means of a unitary operator which corresponds to inhomogeneous rotations of the (effective) atomic spins assigned to the lattice sites of a ferromagnetic crystal. The Hamiltonian was assumed to be quadratic with respect to spin operators (second-order interactions). Linear terms corresponding to external magnetic field were not considered. Neither the interaction tensors nor the crystal lattice were specified, and no restriction was imposed on the range of the interaction between the lattice spins. Under these assumptions it was shown that the mapping of the Hamiltonian from the space of Bloch's physical spin waves onto the space of Dyson's ideal spin waves can be carried through quite easily, by means of a simple substitution procedure (see also [8]) which was proven to be applicable to any spin Hamilto-

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nian of Heisenberg's type. Diagonalization of the spin wave Hamiltonian within Bloch's linear approximation can always be achieved by employing Bogolyubov's general transformation [6] to the ideal spin wave creation and destruction operators. Thus, it has been shown that all the specifications and simplifications of the problem that are usually made at the very beginning can be deferred until one has to calculate thermodynamic quantities (such as the partition function, free energy, magnetic susceptibility), including the eventual choice of the most suitable spin wave representation.

As for the energy spectrum, a set of general equations has been derived which are to be specified and solved in each particular case. These equations actually represent the conditions under which the (approximate) diagonalization of the Hamiltonian is automatically ensured. A detailed examination [1a] of those conditions shows, *inter alia*, that the application of the spin wave formalism in its present form to hexagonal crystal lattices is admissible, which justifies the employment of the spin wave method to the very important group of uniaxial ferromagnets. On the other hand, the anisotropic effects of cubic ferromagnetic lattices to be counted for require in the Heisenberg approach a Hamiltonian with at least quartic couplings between the lattice spins [9]. It thus appears that the formalism proposed in [1a] to be applicable to real ferromagnets has to be extended to interactions of fourth order whilst the presence of external magnetic field requires linear terms to be added. It is precisely this extension which is the main purpose of the present paper, the other assumptions as well as the mathematical approach being essentially the same as in [1a]. In particular, no specifications will be made about the crystal lattice and the form of the interaction tensors, nor the interaction limited to any particular neighbourhood. The linear Bloch approximation will also be retained, that is, interactions between the (ideal) spin waves are being neglected. The justification of this restriction resides partly in the fact that those interactions have usually no (or very little) influence on the diagonalization of the bilinear part of the Hamiltonian, and partly in the supposition that Dyson's proof of the negligibility of these interactions in the isotropic case is very likely to be generally valid.

Within these limitations we show also the approaches of Dyson [2], Charap and Weiss [3], and Szaniecki [4] to be particular cases following easily from our general formalism, under proper specifications and simplifications.

While merely indicating major steps in our calculations, we shall frequently refer to [1a] where all the details can be found.

## 2. Diagonalization of the Hamiltonian

We start with the Hamiltonian

$$H = \sum_j L^a S_j^a + \sum_{jk} (H_{jk}^{ab} S_j^a S_k^b + Q_{jk}^{abcd} S_j^a S_j^b S_k^c S_k^d), \quad (1)$$

where  $a, b, c, d$  ( $= 1, 2, 3$ ) are tensor indices, and  $j, k$  denote the spatial *vectors* pointing to the corresponding lattice sites. (To simplify the notation we shall not use arrows above lattice vector symbols). For tensor indices Einstein summation convention is adopted throughout the paper.

To avoid self-interaction we put  $H_{jj} = Q_{jj} = 0$ . In the Zeeman term  $L^a = g|\mu_B|\mathcal{H}^a$ , where  $g$  denotes Lande's factor,  $\mu_B$  — Bohr's magneton and  $\mathcal{H} = (\mathcal{H}^a\mathcal{H}^a)^{1/2}$  is the external magnetic field strength.

$H_{jk}^{ab}$  and  $Q_{jk}^{abcd}$  stand for the interaction tensors of the second and fourth rank, respectively, and the only restriction we impose on them has the form

$$\begin{aligned} H_{jk}^{ab} &= H_{kj}^{ab} = H_{kj}^{ba}, \\ Q_{jk}^{abcd} &= Q_{jk}^{bacd} = Q_{jk}^{badc} = Q_{jk}^{abdc} = Q_{kj}^{abcd} = Q_{kj}^{bacd}. \end{aligned} \quad (2)$$

The spin operators  $S_j^a$  satisfy the well-known commutation rules

$$[S_j^a, S_k^b] = i\delta_{jk}\varepsilon^{abc}S_j^c, \quad (3)$$

where  $\varepsilon^{abc}$  is the antisymmetric unit pseudo-tensor.

One can define the operators  $S_j^\pm$  by the relation

$$S_j^\pm = S_j^1 \pm iS_j^2. \quad (4)$$

Commutation rules for these operators follow easily from (2) and (3), and are of the form

$$[S_j^+, S_k^-] = 2\delta_{jk}S_j^3, \quad [S_j^-, S_k^3] = \delta_{jk}S_j^-, \quad [S_j^3, S_k^+] = \delta_{jk}S_j^+. \quad (5)$$

These operators act in the space of simultaneous eigenvectors of the operators  $S_j^3$  where the ground state of an isotropic ferromagnet is defined by [1, 1a, 2]

$$S_j^3|0\rangle = -S|0\rangle; \quad S_j^-|0\rangle = \langle 0|S_j^+ = 0, \quad (6)$$

$S$  being the maximum eigenvalue of  $S_j^3$ .

One can generate an arbitrary eigenstate  $|u\rangle$  of the operators  $S_j^3$  by applying repeatedly the operators (3) to the ground state (5)

$$|\dots u_j \dots\rangle \equiv |u\rangle = \left\{ \prod_j \left[ \frac{(2S-u_j)!}{(2S)!(u_j)!} \right]^{1/2} (S_j^+)^{u_j} \right\} |0\rangle, \quad (7)$$

$u_j$  being a positive integer. This is an orthonormal and complete set of  $(2S+1)^N$  independent vectors where  $N$  is the number of lattice sites. One can easily verify that

$$0 \leq u_j \leq 2S. \quad (8)$$

The states (7) however, are not eigenstates of the isotropic Hamiltonian, much less so for a Hamiltonian of the form (1). None the less, in the isotropic case (*i. e.*,  $H_{jk}^{ab} = H_{jk}\delta^{ab}$  and  $Q_{jk}^{abcd} = 0$ ) the use of the representation (7) is reasonably justified, at least in two limiting cases when either the external (uniform) magnetic field is sufficiently strong or the temperature much lower than the respective Curie temperature. This is so because in the first case one can take advantage of the fact that the operator  $\sum_j S_j^3$  is diagonal in the representation (7)

and commutes with the isotropic part of the Hamiltonian, and in the second case one still has the argument that  $|0\rangle$  is the ground state of the Hamiltonian. It is the latter argument which fails in the anisotropic case. For many reasons which were given in [1a] it seems suit-

able to widen the representation (7) by introducing the unitary operators  $U_j$  causing rotations of the spin vectors  $S_j^a$  by the angles  $\varphi_j$  around rotation axes with direction cosines  $e_j^a$ . These operators have the form

$$U_j = \exp \{i\varphi_j e_j^a S_j^a\}, \quad \prod_j U_j \equiv U. \quad (9)$$

This is equivalent to introducing the rotation matrices  $R_j^{ab}$  defined as

$$U_j S_j^a U_j^\dagger = R_j^{ab} S_j^b, \quad (10)$$

where

$$R_j^{ab} = \varepsilon^{acb} e_j^c \sin \varphi_j + \delta^{ab} \cos \varphi_j + e_j^a e_j^b (1 - \cos \varphi_j) \quad (11)$$

(cf. Ref. [14] in [1a]).

The operators (9) when applied to the states (7) give the class of representations

$$|v\rangle = U^\dagger |u\rangle. \quad (12)$$

Since  $U$  is a unitary transformation, it is obvious that the vectors  $|v\rangle$  are orthonormal.

It was shown in [1a] that instead of working with the representation  $|v\rangle$  one can transform the Hamiltonian and further on use the standard representation (7). When applying the transformation (9) to the Hamiltonian (1) and utilizing Eq. (10) one obtains

$$UHU^\dagger \equiv \tilde{H} = \sum_j \tilde{L}_j^b S_j^b + \sum_k (A_{jk}^{ab} S_j^a S_k^b + \tilde{Q}_{jk}^{abcd} S_j^a S_j^b S_k^c S_k^d), \quad (13)$$

where

$$L_j^b \equiv L^a R_j^{ab}, \quad A_{jk}^{ab} = H_{jk}^{cd} R_j^{ca} R_k^{db}, \quad \tilde{Q}_{jk}^{abcd} = Q_{jk}^{efgh} R_j^{ea} R_j^{fb} R_k^{gc} R_k^{hd}. \quad (14)$$

The coefficients  $A_{jk}^{ab}$  and  $\tilde{Q}_{jk}^{abcd}$  have the properties

$$A_{jk}^{ab} = A_{kj}^{ba} \quad (15)$$

which follow from Eqs (2) and (11), and

$$\tilde{Q}_{jk}^{abcd} = \tilde{Q}_{jk}^{bacd} = \tilde{Q}_{jk}^{badc} = \tilde{Q}_{jk}^{abdc} \quad (16)$$

which implies

$$(Q_{jk}^{abcd} R_j^{am} R_j^{bn}) R_k^{cs} R_k^{dt} = (Q_{jk}^{abcd} R_j^{bm} R_j^{an}) R_k^{cs} R_k^{dt}. \quad (17)$$

The next step consists in introducing the complete and orthonormal set of oscillatory states

$$|\dots u_j \dots \rangle \equiv |u\rangle \equiv \left\{ \prod_j (u_j!)^{-1/2} (\eta_j^\dagger)^{u_j} \right\} |0\rangle \quad (18)$$

where  $u_j$  is an integer taking values from 0 to  $\infty$ . The notation  $|\cdot\rangle$  is used to distinguish these states from those defined by Eq. (7). The oscillatory operators  $\eta_j$  and  $\eta_j^\dagger$  attached to the lattice sites satisfy the familiar commutation rules

$$[\eta_j, \eta_k^\dagger] = \delta_{jk}, \quad [\eta_j, \eta_k] = [\eta_j^\dagger, \eta_k^\dagger] = 0. \quad (19)$$

Now, it was pointed out in [1] that the easiest way of mapping the Hamiltonian from the space of physical onto the space of ideal spin waves is to pass from the operators (5) to the operators (19), and that this is equivalent to the substitution

$$S_j^+ \rightarrow (2S)^{1/2} \eta_j^+; \quad S_j^- \rightarrow (2S)^{1/2} \left(1 - \frac{\eta_j^+ \eta_j}{2S}\right) \eta_j; \quad S_j^3 \rightarrow -S + \eta_j^+ \eta_j \quad (20)$$

(see also [8]) which corresponds to the transition (i)  $\rightarrow$  (J) in [1]. When applying this substitution to the Hamiltonian (13) and neglecting all terms involving more than two operators (19) one obtains

$$\begin{aligned} \tilde{H} = & \sum_j (-SL_j^2 + L_j^2 \eta_j^+ \eta_j + (2S)^{1/2} L_j^1 \eta_j^+ + (2S)^{1/2} \bar{L}_j^1 \eta_j) + \\ & + \sum_{jk} \left\{ S^2 (M_{jk}^0 + B_{jk}^4) - (2S)^{1/2} S (B_{jk}^3 + M_{jk}^1) \eta_j^+ - (2S)^{1/2} S (\bar{B}_{jk}^3 + \bar{M}_{jk}^1) \eta_j + \right. \\ & + \frac{1}{2} S (B_{jk}^1 + M_{jk}^3) \eta_j^+ \eta_k^+ + \frac{1}{2} S (\bar{B}_{jk}^1 + \bar{M}_{jk}^3) \eta_j \eta_k + S (B_{jk}^2 + M_{jk}^5) \eta_j^+ \eta_k + \\ & \left. + S (M_{jk}^4 - 2B_{jk}^4) \eta_j^+ \eta_j + \frac{1}{2} S M_{jk}^2 (\eta_j^+)^2 + \frac{1}{2} S \bar{M}_{jk}^2 (\eta_j)^2 \right\}, \quad (21) \end{aligned}$$

where

$$\begin{aligned} L_j^1 &= \frac{1}{2} (\tilde{L}_j^1 - i\tilde{L}_j^2), \quad L_j^2 = \tilde{L}_j^3, \\ B_{jk}^1 &= A_{jk}^{11} - A_{jk}^{22} - i(A_{jk}^{12} + A_{jk}^{21}), \quad B_{jk}^2 = A_{jk}^{11} + A_{jk}^{22}, \\ B_{jk}^3 &= (A_{jk}^{13} - iA_{jk}^{23}) + (A_{jk}^{31} - iA_{jk}^{32}), \quad B_{jk}^4 = A_{jk}^{33}, \end{aligned} \quad (22)$$

and  $M_{jk}^r$  ( $r = 0, \dots, 5$ ) are rather lengthy combinations of  $\tilde{Q}_{jk}^{abcd}$  which are listed in the Appendix.

By carrying out the Fourier transformation

$$\eta_j^+ = N^{-1/2} \sum_{\lambda} \alpha_{\lambda}^+ e^{-i\lambda j} \quad (23)$$

we pass to the reciprocal lattice with lattice vectors  $\lambda$ . (Here we again drop the arrows above vector symbols. Note that  $\lambda j$  is thus the scalar product of two vectors).

The operators  $\alpha_{\lambda}, \alpha_{\lambda}^+$  obey commutation rules which may be obtained directly from Eqs (23) and (19):

$$\begin{aligned} [\alpha_{\lambda}, \alpha_{\mu}^+] &= \delta_{\lambda\mu}, \quad [\alpha_{\lambda}, \alpha_{\mu}] = [\alpha_{\lambda}^+, \alpha_{\mu}^+] = 0, \\ \alpha_{\lambda}^+ \alpha_{\lambda} &= a_{\lambda}, \quad a_{\lambda} = 0, 1, 2, \dots \end{aligned} \quad (24)$$

Here  $a_{\lambda}$  can be interpreted as the number of spin waves having the wave vector  $\lambda$ , the corresponding (orthonormal) states being defined as

$$|\dots a_{\lambda} \dots \gg \equiv |a \gg = \left\{ \prod_{\lambda} (a_{\lambda}!)^{-1/2} (\alpha_{\lambda}^+)^{a_{\lambda}} \right\} |0 \gg. \quad (25)$$

Transformation (23) applied to the Hamiltonian (21) provides

$$\begin{aligned}\tilde{H} = & \sum_{\lambda\mu} \{SN[S(M_{\lambda\mu}^0 + B_{\lambda\mu}^4) - L_{\lambda+\mu}^2] \delta_{\lambda,0} \delta_{\mu,0} - \\ & - (2SN)^{1/2} \delta_{\mu,0} [S(B_{\lambda\mu}^3 + M_{\lambda\mu}^1) - L_{\lambda+\mu}^1] \alpha_{\lambda}^+ - \\ & - (2SN)^{1/2} \delta_{\mu,0} [S(\bar{B}_{\lambda\mu}^3 + \bar{M}_{\lambda\mu}^1) - \bar{L}_{\lambda+\mu}^1] \alpha_{\lambda} + \\ & + \frac{1}{2} S[B_{\lambda\mu}^1 + M_{\lambda+\mu,0}^2 + M_{\lambda,\mu}^3] \alpha_{\lambda}^+ \alpha_{\mu}^+ + \frac{1}{2} S[\bar{B}_{\lambda,\mu}^1 + \bar{M}_{\lambda+\mu,0}^2 + \bar{M}_{\lambda,\mu}^3] \alpha_{\lambda} \alpha_{\mu} + \\ & + [S(B_{-\lambda,\mu}^2 - 2B_{\mu-\lambda,0}^4 + M_{\lambda-\mu,0}^4 + M_{\lambda,-\mu}^5) + L_{\lambda-\mu}^2] \alpha_{\lambda}^+ \alpha_{\mu},\end{aligned}\quad (26)$$

where the coefficients  $B_{\lambda,\mu}^q$  ( $q = 1, 2, 3, 4$ ) and  $M_{\lambda,\mu}^r$  ( $r = 0, 1, \dots, 5$ ) are the Fourier transforms of  $B_{j,k}^q$  and  $M_{j,k}^r$ , respectively (see Eq. (22) and Appendix), and

$$L_{\lambda+\mu}^t = 1/N \sum_j L_j^t \exp \{-ij(\lambda+\mu)\}, \quad t = 1, 2 \quad (27)$$

with  $L_j^t$  defined by Eq. (22).

It is useful to introduce the abbreviations

$$\begin{aligned}P_{\lambda,\mu}^0 &= M_{\lambda,\mu}^0, & P_{\lambda,\mu}^1 &= M_{\lambda,\mu}^1, & P_{\lambda,\mu}^2 &= M_{\lambda+\mu,0}^2 + M_{\lambda,\mu}^3 \\ P_{\lambda,\mu}^3 &= M_{\lambda-\mu,0}^4 + M_{\lambda,-\mu}^5\end{aligned}\quad (28)$$

and

$$\begin{aligned}W_{\lambda,\mu}^0 &= \frac{1}{4} (P_{\lambda,\mu}^0 + B_{\lambda,\mu}^4) - \frac{1}{4S} L_{\lambda+\mu}^2, & W_{\lambda,\mu}^1 &= \frac{1}{2} (B_{\lambda,\mu}^3 + P_{\lambda,\mu}^1) - \frac{1}{2S} L_{\lambda+\mu}^1, \\ W_{\lambda,\mu}^2 &= \frac{1}{4} (B_{\lambda,\mu}^1 + P_{\lambda,\mu}^2), & W_{\lambda,\mu}^3 &= \frac{1}{2} (B_{-\lambda,\mu}^2 - 2B_{\mu-\lambda,0}^4 + P_{\lambda,\mu}^3) + \frac{1}{2S} L_{\lambda-\mu}^2.\end{aligned}\quad (29)$$

Hence, the Hamiltonian (26) takes the form

$$\begin{aligned}H = 2S \sum_{\lambda\mu} \{ & (2SN) W_{\lambda,\mu}^0 \delta_{\lambda,0} \delta_{\mu,0} - (2SN)^{1/2} [W_{\lambda,\mu}^1 \alpha_{\lambda}^+ + \bar{W}_{\lambda,\mu}^1 \alpha_{\lambda}] \delta_{\mu,0} + \\ & + [W_{\lambda,\mu}^2 \alpha_{\lambda}^+ \alpha_{\mu}^+ + \bar{W}_{\lambda,\mu}^2 \alpha_{\lambda} \alpha_{\mu}] + W_{\lambda,\mu}^3 \alpha_{\lambda}^+ \alpha_{\mu} \}.\end{aligned}\quad (30)$$

To eliminate the terms linear in the operators  $\alpha_{\lambda}$ ,  $\alpha_{\lambda}^+$  we use the "shifting" transformation

$$\alpha_{\lambda} = b_{\lambda} + \beta_{\lambda}. \quad (31)$$

Insertion of (31) into the Hamiltonian (30) yields

$$\begin{aligned}\tilde{H} = 2S(2SN) E_0 + 2S(2SN)^{1/2} \sum_{\lambda} [C_{\lambda}^1 \beta_{\lambda}^+ + \bar{C}_{\lambda}^1 \beta_{\lambda}] + \\ + 2S \sum_{\lambda\mu} [C_{\lambda,\mu}^1 \beta_{\lambda}^+ \beta_{\mu}^+ + C_{\lambda,\mu}^2 \beta_{\lambda} \beta_{\mu} + C_{\lambda,\mu}^0 \beta_{\lambda}^+ \beta_{\mu}],\end{aligned}\quad (32)$$

where

$$E_0 = \frac{1}{4} \sum_{\lambda\mu} W_{\lambda,\mu}^0 - \frac{1}{2} (2SN)^{-1} \sum_{\lambda\mu} [W_{\lambda,\mu}^1 \bar{b}_\lambda + \bar{W}_{\lambda,\mu}^1 b_\lambda] + \\ + \frac{1}{4} (2SN)^{-1} \sum_{\lambda\mu} [W_{\lambda,\mu}^2 \bar{b}_\lambda \bar{b}_\mu + \bar{W}_{\lambda,\mu}^2 b_\lambda b_\mu] + 4 \sum_{\lambda\mu} W_{\lambda,\mu}^3 \bar{b}_\mu b_\lambda, \quad (33)$$

$$C_\lambda^1 = -\frac{1}{2} \sum_{\mu} W_{\lambda,\mu}^1 + (2SN)^{-1} \sum_{\mu} \left[ \frac{1}{4} (W_{\lambda,\mu}^2 + W_{\mu,\lambda}^2) \bar{b}_\mu + W_{\mu,\lambda}^3 \bar{b}_\mu \right], \quad (34)$$

$$C_{\lambda,\mu}^1 = \frac{1}{4} W_{\lambda,\mu}^2 = \bar{C}_{\lambda,\mu}^2, \quad (35)$$

$$C_{\lambda,\mu}^0 = W_{\lambda,\mu}^3. \quad (36)$$

To determine the constants  $b_\lambda$  one has to solve the set of linear algebraic equations

$$C_\lambda^1 = 0 \quad (37)$$

Had we not imposed the condition (17), the Hamiltonian (32) would not be self-adjoint, and in consequence instead of  $N$  equations (37) for the constants  $b_\lambda$  we would have  $2N$  equations

$$C_\lambda^1 = 0, \quad C_\lambda^2 = 0 \quad (38)$$

It would be therefore necessary to introduce the consistency condition

$$C_\lambda^1 = \bar{C}_\lambda^2 \quad (39)$$

restricting rather strongly the class of permissible rotations (9). After solving Eqs (37) one can employ the general transformation of Bogolyubov [6] by passing to new Bose operators  $\gamma_e, \gamma_e^+$

$$\beta_\lambda = \sum_e (u_{\lambda e} \gamma_e + \bar{v}_{\lambda e} \gamma_e^+) \quad (40)$$

These operators will diagonalize the Hamiltonian (32) provided the following conditions are fulfilled (see also [7]):

$$C_{\lambda,\mu}^1 = \bar{C}_{\lambda,\mu}^2, \quad C_{\lambda,\mu}^0 = \bar{C}_{\mu,\lambda}^0. \quad (41)$$

As in [1a] conditions (41) are here automatically satisfied.

The situation is much the same as in [1a], *i. e.* the neglect of higher order terms in the Hamiltonian (30) does not change the conditions (41) as long as the linear terms in (30) vanish automatically, that is if

$$W_{\lambda,0}^1 = 0, \quad (42)$$

as the transformation (31) produces contributions to the bilinear terms following from the interactions of higher order.

For the Hamiltonian (1) condition (42) has the form

$$\begin{aligned}
 & \left( S - \frac{1}{2} \right) \sum_j Q_{jk}^{abcd} \left[ (R_j^{a3} R_j^{b3} R_k^{c3} R_k^{d3} + R_j^{a3} R_j^{b1} R_k^{c3} R_k^{d3}) S + \right. \\
 & \quad + \frac{1}{2} (R_j^{a1} R_j^{b1} R_k^{c1} R_k^{d3} + R_j^{a1} R_j^{b3} R_k^{c1} R_k^{d1} + R_j^{a1} R_j^{b3} R_k^{c2} R_k^{d2} + \\
 & \quad \left. + R_j^{a2} R_j^{b2} R_k^{c1} R_k^{d3}) \right] + \sum_j H_{jk}^{ab} (R_j^{a1} R_k^{b3} + R_j^{a3} R_k^{b1}) - \frac{1}{2S} \sum_j L^a R_j^{a1} \\
 & = \sum_j Q_{jk}^{abcd} \left[ (R_j^{a1} R_j^{b1} R_k^{c2} R_k^{d3} + R_j^{a2} R_j^{b3} R_k^{c1} R_k^{d1} + R_j^{a2} R_j^{b2} R_k^{c2} R_k^{d3} + \right. \\
 & \quad \left. + R_j^{a2} R_j^{b3} R_k^{c2} R_k^{d2}) \left( S - \frac{1}{2} \right) + (R_j^{a3} R_j^{b3} R_k^{c3} R_k^{d2} + R_j^{a3} R_j^{b2} R_k^{c3} R_k^{d3}) \frac{1}{2} S \right] + \\
 & \quad + \sum_j H_{jk}^{ab} (R_j^{a2} R_k^{b3} + R_j^{a3} R_k^{b2}) - \frac{1}{2S} \sum_j L^a R_j^{a2} = 0 \quad (43)
 \end{aligned}$$

as can be deduced from Eqs (29), (28), (22), (14) and Appendix.

Substituting the transformation (40) into the Hamiltonian (32) and taking into account Eqs (37) and (41) one has

$$\tilde{H} = 2S(2SN) E_0 + 2S \sum_{\mathbf{e}} D_{\mathbf{e}} + 2S \sum_{\mathbf{e}\sigma} [D_{\mathbf{e}\sigma}^1 \gamma_{\mathbf{e}}^+ \gamma_{\sigma}^+ + \bar{D}_{\mathbf{e}\sigma}^1 \gamma_{\mathbf{e}} \gamma_{\sigma}] + 2S \sum_{\mathbf{e}\sigma} D_{\mathbf{e}\sigma}^0 \gamma_{\mathbf{e}}^+ \gamma_{\sigma}, \quad (44)$$

where

$$D_{\mathbf{e}} = \sum_{\lambda\mu} [C_{\lambda,\mu}^1 v_{\lambda\mathbf{e}} \bar{u}_{\mu\mathbf{e}} + \bar{C}_{\lambda,\mu}^1 u_{\lambda\mathbf{e}} \bar{v}_{\mu\mathbf{e}} + C_{\lambda\mu}^0 v_{\lambda\mathbf{e}} v_{\mu\mathbf{e}}], \quad (45)$$

$$D_{\mathbf{e}\sigma}^1 = \sum_{\lambda\mu} [C_{\lambda,\mu}^1 \bar{u}_{\lambda\mathbf{e}} \bar{u}_{\mu\sigma} + \bar{C}_{\lambda,\mu}^1 \bar{v}_{\lambda\mathbf{e}} \bar{v}_{\mu\sigma} + C_{\lambda\mu}^0 \bar{u}_{\lambda\mathbf{e}} \bar{v}_{\mu\sigma}]. \quad (46)$$

$$D_{\mathbf{e}\sigma}^0 = \sum_{\lambda\mu} [2C_{\lambda,\mu}^1 \bar{u}_{\lambda\mathbf{e}} v_{\mu\sigma} + 2\bar{C}_{\lambda,\mu}^1 \bar{v}_{\lambda\mathbf{e}} u_{\mu\sigma} + C_{\lambda,\mu}^0 \bar{u}_{\lambda\mathbf{e}} u_{\mu\sigma} + \bar{C}_{\lambda,\mu}^0 \bar{v}_{\lambda\mathbf{e}} v_{\mu\sigma}]. \quad (47)$$

As the operators  $\gamma_{\mathbf{e}}, \gamma_{\mathbf{e}}^+$  and  $\beta_{\lambda}, \beta_{\lambda}^+$  obey identical commutation rules, we have

$$\begin{aligned}
 \sum_{\mathbf{e}} (u_{\lambda\mathbf{e}} \bar{u}_{\mu\mathbf{e}} - \bar{v}_{\lambda\mathbf{e}} v_{\mu\mathbf{e}}) &= \delta_{\lambda\mu}, \\
 \sum_{\mathbf{e}} (u_{\lambda\mathbf{e}} \bar{v}_{\mu\mathbf{e}} - \bar{v}_{\lambda\mathbf{e}} u_{\mu\mathbf{e}}) &= 0,
 \end{aligned} \quad (48)$$

and, from the reverse transformation [7],

$$\begin{aligned}
 \sum_{\lambda} (\bar{u}_{\lambda\mathbf{e}} u_{\lambda\sigma} - \bar{v}_{\lambda\mathbf{e}} v_{\lambda\sigma}) &= \delta_{\mathbf{e}\sigma}, \\
 \sum_{\lambda} (u_{\lambda\mathbf{e}} \bar{v}_{\lambda\sigma} - \bar{v}_{\lambda\mathbf{e}} u_{\lambda\sigma}) &= 0.
 \end{aligned} \quad (49)$$



Thus to diagonalize the Hamiltonian (44) one has to solve the equations

$$D_{e\sigma}^1 = 0, \quad D_{e\sigma}^0 = E_e \delta_{e\sigma}, \quad (50)$$

or the equivalent set of linear equations [6,7]

$$\begin{aligned} E_\sigma u_{\lambda\sigma} &= \sum_\mu (2C_{\lambda\mu}^1 v_{\mu\sigma} + C_{\lambda\mu}^0 u_{\mu\sigma}), \\ -E_\sigma v_{\lambda\sigma} &= \sum_\mu (2\bar{C}_{\lambda\mu}^1 u_{\mu\sigma} + \bar{C}_{\lambda\mu}^0 v_{\mu\sigma}). \end{aligned} \quad (51)$$

In this way the Hamiltonian (44) becomes

$$\tilde{H} = 2SE'_0 + 2S \sum_e E_e \gamma_e^+ \gamma_e, \quad (52)$$

where

$$E'_0 = 2SN E_0 + \sum_e D_e. \quad (53)$$

### 3. Correspondence to other authors

We shall show briefly what specifications and simplifications have to be made to obtain the spin wave Hamiltonians considered by Dyson [2], Charap and Weiss [3] and Szaniecki [4]. In all those papers cubic crystal lattices were considered and the interactions restricted to the nearest neighbours. The external magnetic field was taken into account, the remaining part of the Hamiltonian being isotropic in [2] and anisotropic in [3, 4].

In [3], the anisotropic part was expressed in terms of pseudo-dipolar interactions, and in [4] quadrupolar interactions were included. In all three papers the standard representation (7) has been used. The linear part of the Hamiltonians which in [2] was automatically diagonal has in [3] and [4] not been fully diagonalized. Although in the latter cases the diagonalization can easily be carried out by means of a rather simple transformation of the type (40), we will show the correspondence to the nondiagonalized Hamiltonians as used by those authors, in order to make the comparison of the respective expressions easier. Our notation differs slightly, but not significantly from that used in those papers.

Dyson's [2] assumptions are as follows: isotropic interactions of the second order, homogeneous external magnetic field parallel to the  $x_3$ -axis, nearest neighbours interaction, cubic crystal lattice (s. c., b. c. c., and f. c. c.), standard representation (7). Denoting by  $\Delta$  the nearest neighbour lattice vectors we thus have:

$$\begin{aligned} k &= j + \Delta, \quad R_j^{ab} = R_k^{ab} = \delta^{ab} \\ H_{jk}^{ab} &\equiv H_{j\Delta}^{ab} = -\frac{1}{2} J \delta^{ab}, \quad L^a = L \delta^{a3}, \\ Q_{jk}^{abcd} &= \tilde{Q}_{jk}^{abcd} = M_{jk}^r = M_{\lambda\mu}^r = P_{\lambda\mu}^s = 0 \\ & (r = 0, \dots, 5); \quad (s = 0, 1, 2, 3) \end{aligned} \quad (54)$$

(see Eqs (1), (14), (26), (28) and Appendix). Hence, from (14) and (22) we obtain

$$\begin{aligned} B_{jk}^1 &= B_{jk}^3 = 0, & B_{jk}^2 &= -J, \\ B_{jk}^4 &= -\frac{1}{2}J, & L_j^1 &= 0, & L_j^2 &= L, \end{aligned} \quad (55)$$

and in the reciprocal lattice, after the transformation (23),

$$\begin{aligned} B_{\lambda\mu}^1 &= B_{\lambda\mu}^3 = 0, & B_{\lambda\mu}^2 &= -J\delta(\lambda+\mu)\gamma_\mu, \\ B_{\lambda\mu} &= -J\delta(\lambda+\mu)\gamma_\mu, & L_{\lambda+\mu} &= L\delta(\lambda+\mu), \end{aligned} \quad (56)$$

where

$$\gamma_\mu = \sum_{\Delta} e^{i\mu\Delta}. \quad (57)$$

By inserting (54) into the Hamiltonian (30) we get, because of Eq. (29),

$$H = -LSN - \frac{1}{2}JNS^2\gamma_0 + \sum_{\lambda} (L + \varepsilon_{\lambda})\alpha_{\lambda}^{\dagger}\alpha_{\lambda}, \quad (58)$$

where

$$\varepsilon_{\lambda} = JS(\gamma_0 - \gamma_{\lambda}). \quad (59)$$

This is exactly the spin wave energy spectrum obtained by Dyson.

Charap and Weiss [3] made the following assumptions: isotropic interactions of Heisenberg type, anisotropic interactions of pseudo-dipolar form, uniform external magnetic field, nearest neighbour approximation, cubic crystal lattice, standard representation (7) and  $S = \frac{1}{2}$ . Hence,

$$\begin{aligned} k &= j + \Delta, & R_j^{ab} &= R_k^{ab} = \delta^{ab}, \\ H_{jk}^{ab} &\equiv H_{j\Delta}^{ab} = -J\delta^{ab} + 2\varepsilon\Delta^a\Delta^b, & L^a &= L\delta^{a3}, \\ Q_{jk}^{abcd} &= \tilde{Q}_{jk}^{abcd} = M_{jk}^r = M_{\lambda\mu}^r = P_{\lambda\mu}^s = 0, \end{aligned} \quad (60)$$

where  $\Delta^a$  ( $a = 1, 2, 3$ ) are the components of  $\Delta$ .

With  $\Delta^{\pm}$  defined as

$$\Delta^{\pm} = \Delta^1 \pm i\Delta^2 \quad (61)$$

one has because of Eqs (14) and (22)

$$\begin{aligned} B_{\lambda\mu}^1 &= 2\varepsilon\delta(\lambda+\mu) \sum_{\Delta} (\Delta^+)^2 e^{i\mu\Delta}, \\ B_{-\lambda,\mu}^2 &= -2J\delta(\lambda-\mu) \sum_{\Delta} e^{i\mu\Delta} + 2\varepsilon\delta(\lambda-\mu) \sum_{\Delta} e^{i\mu\Delta} (\Delta^+ \Delta^-), \\ B_{\lambda,\mu}^3 &= 2\varepsilon\delta(\lambda+\mu) \sum_{\Delta} e^{i\mu\Delta} (\Delta^+ \Delta^3), \\ B_{\mu-\lambda,0}^4 &= -J \sum_{\Delta} \delta(\lambda-\mu) + 2\varepsilon \sum_{\Delta} (\Delta^3)^2 \delta(\lambda-\mu), \\ B_{\lambda,\mu}^4 &= -J\delta(\lambda+\mu)\gamma_{\mu} + 2\varepsilon\delta(\lambda+\mu) \sum_{\Delta} e^{i\mu\Delta} (\Delta^3)^2, \\ L_j^2 &= L, & L_j^1 &= 0. \end{aligned} \quad (62)$$

The insertion of (62) into (29) and (30) yields

$$\begin{aligned}
 H = & -\frac{1}{4} JN\gamma_0 + \frac{1}{2} \varepsilon N \sum_{\Delta} (\Delta^3)^2 - \frac{1}{2} NL + \\
 & + \frac{1}{2} \varepsilon \sum_{\mu} [e^{i\mu\Delta} (\Delta^+)^2 \alpha_{\mu}^+ \alpha_{-\mu}^+ + e^{-i\mu\Delta} (\Delta^-)^2 \alpha_{\mu} \alpha_{-\mu}] - \\
 & - J \sum_{\mu\Delta} (e^{i\mu\Delta} - 1) \alpha_{\mu}^+ \alpha_{\mu} + \sum_{\mu} L \alpha_{\mu}^+ \alpha_{\mu} + \\
 & + \varepsilon \sum_{\mu\Delta} [e^{i\mu\Delta} \Delta^+ \Delta^- - 2(\Delta^3)^2] \alpha_{\mu}^+ \alpha_{\mu}.
 \end{aligned} \quad (63)$$

The terms linear with respect to  $\alpha_{\lambda}$  and  $\alpha_{\lambda}^+$  in Eq. (30) vanish because of the cubic symmetry. Rearranging (63) we get

$$\begin{aligned}
 H = & -\frac{1}{2} LN - \frac{1}{4} JN\gamma_0 + \frac{1}{2} N\varepsilon \sum_{\Delta} (\Delta^3)^2 + \\
 & + \sum_{\mu} \{L + J \sum_{\Delta} (1 - \cos \vec{\mu}\vec{\Delta}) - \varepsilon \sum_{\Delta} [2(\Delta^3)^2 - (\Delta^+ \Delta^-) \cos \vec{\mu}\vec{\Delta}]\} \alpha_{\mu}^+ \alpha_{\mu} + \\
 & + \frac{1}{2} \varepsilon \sum_{\mu} (v_{\mu} \alpha_{\mu}^+ \alpha_{-\mu}^+ + \bar{v}_{\mu} \alpha_{\mu} \alpha_{-\mu}),
 \end{aligned} \quad (64)$$

where

$$v_{\mu} = \sum_{\Delta} (\Delta^+)^2 e^{i\mu\Delta}.$$

Eq. (64) is identical with the bilinear part of the Hamiltonian used by Charap and Weiss.

The assumptions made by Szaniecki [4] are as follows: cubic crystal lattice (s. c. and b. c. c.), nearest neighbour approximation, isotropic pseudo-dipolar and quadrupolar interactions, uniform external magnetic field, standard representation (7). This corresponds to

$$\begin{aligned}
 k &= j + \Delta, \quad R^{ab} = R_k^{ab} = \delta^{ab}, \\
 H_{jk}^{ab} &\equiv H_{j\Delta}^{ab} = -J\delta^{ab} + 1/2D \left[ \delta^{ab} - \frac{3\Delta^a \Delta^b}{(\Delta)^2} \right], \quad L^a = L\delta^{a3}, \\
 Q_{jk}^{abcd} &\equiv Q_{k\Delta}^{abcd} = \frac{1}{2} Q \Delta^{-4} (\Delta_a \Delta_b \Delta_c \Delta_d).
 \end{aligned} \quad (65)$$

Therefore, by virtue of Eqs (14) and (22) we have

$$\begin{aligned}
 B_{\lambda,\mu}^1 &= -\frac{3}{2} D \sum_{\Delta} e^{i\mu\Delta} (\Delta^+/\Delta)^2 \delta(\lambda+\mu), \\
 B_{\lambda,\mu}^2 &= -2J\delta(\lambda+\mu) \gamma_{\mu} + D\delta(\lambda+\mu) \gamma_{\mu} - 3/2D \sum_{\Delta} e^{i\mu\Delta} \frac{\Delta^+ \Delta^-}{(\Delta^2)} \delta(\lambda+\mu), \\
 B_{\lambda,\mu}^3 &= -3/2D \sum_{\Delta} e^{i\mu\Delta} \frac{\Delta^+ \Delta^3}{(\Delta^2)} \delta(\lambda+\mu),
 \end{aligned}$$

$$B_{\lambda,\mu}^4 = (1/2D - J)\gamma_\mu \delta(\lambda + \mu) - 3/2D \sum_A e^{i\mu A} (A^3/A)^2 \delta(\lambda + \mu),$$

$$L_{\lambda-\mu}^2 = L\delta(\lambda - \mu), \quad L_{\lambda+\mu}^1 = 0, \quad (66)$$

and the quadrupolar coefficients  $P_{\lambda,\mu}^S$  are (see Appendix and Eqs (28))

$$\begin{aligned} P_{\lambda,\mu}^0 = M_{\lambda,\mu}^0 &= \frac{1}{2} Q A^{-4} \delta(\lambda + \mu) \sum_A e^{i\mu A} \left\{ \frac{1}{4} [(A_1)^4 + 2(A_1 A_2)^2 + (A_2)^4] + \right. \\ &\quad \left. + S[(A_1)^2 + (A_2)^2](A_3)^2 + S^2(A_3)^4 \right\}, \\ P_{\lambda,\mu}^1 = M_{\lambda,\mu}^2 &= \frac{1}{2} Q A^{-4} \delta(\lambda + \mu) \sum_A e^{i\mu A} \left\{ 2S^2(A_3)^3 A_1 - S[(A_3)^2 - (A_1)^2 - (A_2)^2] A_1 A_3 - \right. \\ &\quad \left. - \frac{1}{2} [(A_1)^2 + (A_2)^2] A_1 A_3 - i \left( S[(A_1)^2 + (A_2)^2 - (A_3)^2] A_2 A_3 + \frac{1}{2} [(A_1)^2 + (A_2)^2] A_2 A_3 \right) \right\}, \\ P_{\lambda,\mu}^2 = M_{\lambda+\mu,0}^2 + M_{\lambda,\mu}^3 &= \frac{1}{2} Q A^{-4} \delta(\lambda + \mu) \sum_A \{ 2S^2[(A_1)^2 - (A_2)^2](A_3)^2 + S[(A_1)^4 - (A_2)^4] + \\ &\quad + (2S-1)^2(A_3)^2[(A_1)^2 - (A_2)^2] e^{i\mu A} + i(4S^2(A_3)^2 A_1 A_2 + \\ &\quad + 2S[(A_1)^2 + (A_2)^2] A_1 A_2 + (2S-1)^2(A_3)^2 A_1 A_2 e^{i\mu A}) \}, \\ P_{\lambda,\mu}^3 = M_{\lambda-\mu,0}^4 + M_{\lambda,-\mu}^5 &= \frac{1}{2} Q A^{-4} \delta(\lambda - \mu) \sum_A \{ 2S^2[(A_1)^2 + (A_2)^2 - 2(A_3)^2](A_3)^2 + \\ &\quad + S[(A_1)^2 + (A_2)^2]^2 + 2S(A_3)^4 - 3S(A_3)^2[(A_1)^2 + (A_2)^2] - \\ &\quad - \frac{1}{2} [(A_1)^2 + (A_2)^2]^2 + (A_3)^2[(A_1)^2 + (A_2)^2] + \\ &\quad + e^{i\mu A} [(A_1)^2 + (A_2)^2](A_3)^2(2S-1)^2 \}. \end{aligned} \quad (67)$$

Finally, making use of Eqs (19), (30), (66), (67) we have

$$\begin{aligned} H &= -LSN - JNS^2\gamma_0 + QS^2N \left[ \frac{1}{8} \gamma_0 + \frac{1}{6} \gamma_0(S - \frac{1}{2}) + (x_1 + x_2\theta)(S - \frac{1}{2})^2 \right] + \\ &\quad + \sum_\mu \{ \varepsilon_\mu + L + DS(\gamma_\mu - \frac{3}{2}w_\mu) + QS(S - \frac{1}{2})^2[(y_1 + y_2\theta) + z_\mu] \} \alpha_\mu^+ \alpha_\mu + \\ &\quad + \sum_\mu \sum_A e^{i\mu A} \left\{ QS A^{-4} (S - \frac{1}{2})^2 \frac{1}{2} [(A^+)^2 + (A^-)^2](A^3)^2 - \frac{3}{4} SD(A^+/A)^2 \right\} \alpha_\mu^+ \alpha_{-\mu}^+ + \\ &\quad + \sum_\mu \sum_A e^{-i\mu A} \left\{ QS A^{-4} (S - \frac{1}{2})^2 \frac{1}{2} [(A^+)^2 + (A^-)^2](A^3)^2 - \frac{3}{4} SD(A^-/A)^2 \right\} \alpha_\mu \alpha_{-\mu}. \end{aligned} \quad (68)$$

The coefficients  $W_{\lambda\mu}^1$  and  $W_{\lambda\mu}^2$  vanish again because of the cubic symmetry of the crystal lattice. In Eq. (68) we used the notation

$$x_1 = \begin{cases} 1 & \text{for s. c.} \\ 4/9 & \text{for b. c. c.} \end{cases} \quad x_2 = \begin{cases} -2 & \text{for s. c.} \\ 16/9 & \text{for b. c. c.} \end{cases}$$

and

$$\theta = \alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2,$$

Where  $\alpha_1, \alpha_2, \alpha_3$  are direction cosines of the external magnetic field relative to the crystal lattice axes. Furthermore,

$$w_\mu = \Delta^{-2} \sum_{\Delta} (\Delta^+ \Delta^-) e^{i\mu\Delta}, \quad z_\mu = 2/\Delta^4 \sum_{\Delta} (\Delta^+ \Delta^-) (\Delta^3)^2 e^{i\mu\Delta},$$

$$y_1 = \begin{cases} -4 & \text{for s.c.} \\ 0 & \text{for b. c. c.} \end{cases} \quad y_2 = \begin{cases} 12 & \text{for s. c.} \\ -32/3 & \text{for b. c. c.} \end{cases}$$

Eq. (68) corresponds exactly to the bilinear part of Szaniecki's Hamiltonian, though the explicit form of some coefficients was not given in [4].

#### 4. Concluding remarks

The method of dealing with the Hamiltonian including the fourth order interactions, as proposed in this paper, supplements the formalism for the second-order interactions given in [1a]. For the sake of brevity, many steps in the calculations which have been presented in full in [1] and [1a] are only shortly outlined here.

The Hamiltonian we consider here is quite general. Neither the type of the interaction tensors nor the crystal lattice is specified throughout the paper, nor is the interaction limited to any particular neighbourhood. In fact, the only approximation made is the neglect of the contributions of higher terms to the bilinear part of the Hamiltonian (21), which may be produced by the "shifting" transformation (31). If this transformation is not required, *i. e.*, if the coefficients  $W_{\lambda\mu}^1$  and  $W_{\lambda\mu}^2$  in Eq. (30) vanish automatically (as is the case, *e. g.*, in (2. 3. 4)), Eqs (41) are exact conditions for diagonalizing the bilinear part of the ideal spin wave Hamiltonian (30). It is clearly seen that they are related to fourth-order interactions between the lattice spins, and that they impose the symmetry condition (43) on the respective interaction tensors and rotation matrices.

As in [1a], the Hamiltonian (44) can be fully diagonalized, provided the set of linear equations (51) can be solved, and conditions (48) and (49) are satisfied.

The author expresses his gratitude to Dr W. J. Ziętek for suggesting the problem and for valuable discussions during the preparation of this paper.

#### APPENDIX

To simplify the notation we shall use here the abbreviation

$$Q_{jk}^{abcd} \equiv (abcd)$$

hence, the coefficients  $M_{jk}^r$  are of the form

$$\begin{aligned}
 M_{jk}^0 &= S^2 (3333) + \frac{1}{2} S[(3311) + (1133) + (3322) + (2233)] + \\
 &\quad + \frac{1}{4} [(1111) + (1122) + (2211) + (2222)], \\
 M_{jk}^1 &= \left(S - \frac{1}{2}\right) \left\{ S[(3331) + (3133)] + \frac{1}{2} [(1311) + (1113) + (1322) + (2213)] \right\} - \\
 &\quad - i \left(S - \frac{1}{2}\right) [(1123) + (2311) + (2223) + (2322) - (3332) - (3233)], \\
 M_{jk}^2 &= S^2 \{[(3311) + (1133) - (3322) - (2233)] + 2i[(3312) + (1233)]\} + \\
 &\quad + S\{[(1111) - (2222)] + i[(1112) + (1211) + (2122) + (2221)]\}, \\
 M_{jk}^3 &= (2S-1)^2 \{[(3131) - (3232)] + i[(3132) + (2313)]\}, \\
 M_{jk}^4 &= -4S \left(S - \frac{1}{2}\right) (3333) + \left(S - \frac{1}{2}\right) \{(S-1)[(3311) + (1133) + \\
 &\quad + (3322) + (2233)] + [(1111) + (2212) + (1122) + (2222)]\}, \\
 M_{jk}^5 &= (2S-1)^2 [(3131) + (3232)].
 \end{aligned}$$

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