The random phase approximation and crystal field effects in magnetism

1990 J. Phys.: Condens. Matter 2 10435

(http://iopscience.iop.org/0953-8984/2/51/016)

J. Phys.: Condens. Matter **2** (1990) 10435–10450. Printed in the UK

The random phase approximation and crystal field effects in magnetism

G J Bowden and J P D Martin
School of Physics, University of New South Wales, Kensington, NSW 2033, Australia

Received 6 June 1990, in final form 24 August 1990

Abstract. A novel solution to the problem of incorporating a strong axial crystal field interaction into the RPA model of a Heisenberg ferromagnet is presented and discussed. The method is based on a technique widely used in the interpretation of nuclear quadrupole resonance (NQR) experiments, where the Hamiltonian is first transformed into the interaction picture and terms which oscillate rapidly in time are subsequently dropped. It is shown that when this method is applied to an $S = 1$ Heisenberg ferromagnet with an axially symmetric quadrupole interaction, unique solutions can be obtained for the ensemble averages $\langle S_z^n \rangle$. The results are compared with those of earlier workers and it is shown that (i) in the limit $T \rightarrow 0$ K and $D \rightarrow 0$ the usual spin wave result is obtained, (ii) both excitation branches exhibit dispersion at finite temperatures, and (iii) a unique solution can be obtained for the Curie temperature T_C in the presence of crystal fields, in contrast to previous work.

1. Introduction

The problem of incorporating crystal field interactions into the random phase approximation (RPA) for a Heisenberg ferromagnet, has been addressed by many authors. In particular, the reader is referred to the papers of Devlin (1971), Haley and Erdös (1972), Egami and Brooks (1975) and Haley (1978). The central problem, outlined by all the authors, is that of devising a consistent decoupling scheme which will allow ensemble averages to be calculated in a unique manner from the differing Green's functions $\langle\langle A; B \rangle\rangle$ which arise. In particular, the results obtained using Green's function equation of motion for $\langle\langle \hat{T}^1_{\pm 1}(l); \hat{T}^n_{\mp 1}(m) \rangle\rangle$ are not identical to those obtained using $\langle\langle \hat{T}^2_{\pm 1}(l); \hat{T}^n_{\mp 1}(m) \rangle\rangle$. This problem is highlighted once again in section 2 of this paper, using the simple example of a spin $S = 1$ ferromagnet subjected to both Heisenberg exchange and a single-ion axially symmetric quadrupole crystal field interaction.

In this paper, a novel solution to this problen is presented which yields consistent ensemble averages in the presence of a strong axial crystal field. The method, which we shall refer to as the transformed Hamiltonian RPA (T$\mathcal{H}$/RPA), is based on a technique widely used in nuclear quadrupole resonance (NQR) studies. In the interpretation of NQR experiments, the Hamiltonian for the nuclear ensemble is first transformed into the 'interaction picture'. Secondly, terms which oscillate rapidly in time are subsequently dropped to yield a time-independent Hamiltonian (see for example Slichter (1967)). In this paper, it is shown that if this method is applied to the Heisenberg ferromagnet the truncated Hamiltonian gives rise to a consistent decoupling scheme, enabling unique solutions for ensemble averages to be obtained. In addition, it is found that (i) both

0953-8984/90/5110435 + 16 \$03.50

excitation branches (for $S = 1$) exhibit dispersion at finite temperatures, and (ii) in the limit $T \to 0$ K and $D \to 0$, the usual spin wave result is obtained.

2. Heisenberg ferromagnet with an axially symmetric quadratic crystal field: general spin S case

The Hamiltonian in question can be written in the form

$$\mathcal{H} = -\sum_i g\mu_B B_{APP} S_z(i) - \tfrac{1}{2}\sum_{\{i,j\}} \{J_{ij} S(i)\cdot S(j) + K_{ij} S_z(i) S_z(j)\} - \sum_i D \frac{1}{\sqrt{6}} \{3S_z(i)^2 - S(S+1)\} \tag{1}$$

where J_{ij} (K_{ij}) is the isotropic (anisotropic) exchange between the ith and jth atoms and D is the axially symmetric second-order crystal field parameter. Note that for $D > 0$ the easy direction of the magnetisation lies along the z axis. Thus below the Curie temperature T_C, the ensemble averages $\langle S_z^n \rangle$, $n = 1, 2$, etc., are non-zero.

For the purposes of this paper however, we choose to use irreducible tensor operators $\hat{T}_q^n$, in place of the Cartesian operators Sx . . . , because of their superior commutation, construction, contraction and rotational properties. In tensorial form, for general S,

$$\begin{aligned}\mathcal{H} = &- \alpha(S) g\mu_B B_{APP} \left(\sum_i \hat{T}_0^1(i)\right) - \tfrac{1}{2}\sum_{\{i,j\}} \alpha^2(S) J_{ij} \\ &\times (\hat{T}_0^1(i)\hat{T}_0^1(j) - \hat{T}_1^1(i)\hat{T}_{-1}^1(j) - \hat{T}_1^1(i)\hat{T}_1^1(j)) \\ &- \tfrac{1}{2}\sum_{\{i,j\}} \alpha^2(S) K_{ij} \hat{T}_0^1(i)\hat{T}_0^1(j) - \gamma(S) D \left(\sum_i \hat{T}_0^2(i)\right)\end{aligned} \tag{2}$$

where (i) the $\hat{T}_q^n(\alpha)$ are the unit irreducible angular momentum tensor operators (defined for example by Bowden *et al* (1986)) and (ii):

$$\begin{aligned}\alpha(S) &= \left(\frac{(2S+2)!}{12(2S-1)!}\right)^{1/2} \qquad (=\sqrt{2} \text{ for } S = 1) \\ \gamma(S) &= \left(\frac{(2S+3)!}{5!(2S-2)!}\right)^{1/2} \qquad (=1 \text{ for } S = 1)\end{aligned} \tag{3}$$

Following Zubarev (1960), we invoke the double time temperature-dependent Green's function

$$\langle\langle A(t); B(t')\rangle\rangle = -i\theta(t-t')\langle [A(t), B(t')]_- \rangle \tag{4}$$

where A and B are operators, and the other symbols possess their usual meanings. Consequently, on Fourier transforming with respect to $(t - t')$, the Green's function equation of motion

$$E\langle\langle A; B\rangle\rangle = (1/2\pi)\langle [A, B]_- \rangle + \langle\langle [A, \mathcal{H}]_- ; B\rangle\rangle \tag{5}$$

is obtained. For the purposes of this paper we set

$$A = \hat{T}_1^1(l) \qquad B = \hat{T}_{-1}^n(m) \tag{6}$$

in equation (5), for reasons that will become apparent. Secondly, on making use of the commutation relationship

$$[\hat{T}^1_1(l), \hat{T}^n_{q-1}(m)]_- = -\frac{[(n+q)(n-q+1)/2]^{1/2}}{\alpha(S)}\hat{T}^n_q(m)\delta_{lm} \tag{7}$$

we find

$$\begin{aligned}[\hat{T}^1_1(l), \mathcal{H}]_- &= \alpha(S)\Big[\sum_{j\neq l} J_{lj}(\hat{T}^1_1(l)\hat{T}^1_0(j) - \hat{T}^1_0(l)\hat{T}^1_1(j))\Big] \\ &+ \alpha(S)\Big(\sum_{j\neq l} K_{lj}\hat{T}^1_1(l)\hat{T}^1_0(j)\Big) + g\mu_B B_{APP}\hat{T}^1_1(l) \\ &+ \left(\frac{3(2S-1)(2S+3)}{10}\right)^{1/2} D\hat{T}^2_1(l)\end{aligned} \tag{8}$$

for the commutator appearing in the last term in equation (5). Thus the Green's function equation of motion is transformed to

$$\begin{aligned}E\langle\langle\hat{T}^1_1(l); \hat{T}^n_{-1}(m)\rangle\rangle &= a_1[n]\langle\hat{T}^n_0(m)\rangle\delta_{lm}/(2\pi) + g\mu_B B_{APP}\langle\langle\hat{T}^1_1(l); \hat{T}^n_{-1}(m)\rangle\rangle \\ &+ \alpha(S)\sum_{j\neq l}[(J_{lj}+K_{lj})\langle\langle\hat{T}^1_1(l)\hat{T}^1_0(j);\ \hat{T}^n_{-1}(m)\rangle\rangle - J_{lj}\langle\langle\hat{T}^1_0(l)\hat{T}^1_1(j); \hat{T}^n_{-1}(m)\rangle\rangle] \\ &+ \left(\frac{3(2S-1)(2S+3)}{10}\right)^{1/2} D\langle\langle\hat{T}^2_1(l); \hat{T}^n_{-1}(m)\rangle\rangle\end{aligned} \tag{9}$$

where

$$a_1[n] = -\frac{[n(n+1)/2]^{1/2}}{\alpha(S)} \qquad \left(= -\frac{[n(n+1)]^{1/2}}{2} \text{ for } S=1\right). \tag{10}$$

We now invoke the usual random phase approximation (Tyablikov 1959)

$$\begin{aligned}\langle\langle(\hat{T}^1_1(l)\hat{T}^1_0(j) - \hat{T}^1_0(l)\hat{T}^1_1(j)); \hat{T}^n_{-1}(m)\rangle\rangle &\simeq \langle\hat{T}^1_0\rangle \\ &\times(\langle\langle\hat{T}^1_1(l); \hat{T}^n_{-1}(m)\rangle\rangle - \langle\langle\hat{T}^1_1(j); \hat{T}^n_{-1}(m)\rangle\rangle)\end{aligned} \tag{11}$$

in the third term on the right-hand side of equation (8), together with translational invariance. This effectively reduces the three-operator Green's functions appearing in equation (9) to two-operator functions. Thus equation (9) can be re-written in the RPA form

$$\begin{aligned}E\langle\langle\hat{T}^1_1(l); \hat{T}^n_{-1}(m)\rangle\rangle &= a_1[n]\langle\hat{T}^n_0(m)\rangle\delta_{lm}/(2\pi) + g\mu_B B_{APP}\langle\langle\hat{T}^1_1(l); \hat{T}^n_{-1}(m)\rangle\rangle \\ &+ \alpha(S)\langle\hat{T}^1_0\rangle\sum_{j\neq l}[(J_{lj}+K_{lj})\langle\langle\hat{T}^1_1(l); \hat{T}^n_{-1}(m)\rangle\rangle - J_{lj}\langle\langle\hat{T}^1_1(j); \hat{T}^n_{-1}(m)\rangle\rangle] \\ &+ [3(2S-1)(2S+3)/10]^{1/2} D\langle\langle\hat{T}^2_1(l); \hat{T}^n_{-1}(m)\rangle\rangle.\end{aligned} \tag{12}$$

If the quadrupole parameter D is set equal to zero, equation (12) can be solved in the usual manner (see Tahir-Kheli and ter Haar 1962, Bowden *et al* 1986). This procedure involves taking the spatial Fourier transform of equation (12). For nearest-neighbour interactions only, it is easily shown that

$$E(k)G_1 = a_1[n]\langle\hat{T}_0^n\rangle/(2\pi) + [g\mu_B B_{\text{APP}} + \alpha(S)\langle\hat{T}_0^1\rangle\{J(0) + K(0) - J(k)\}]G_1 \tag{13}$$

where (i)

$$G_1 = \sum_m \langle\langle\hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle \exp[-\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_l - \boldsymbol{R}_m)] \tag{14}$$

and (ii)

$$J(k) = \sum_j J_{lj} \exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{\delta}_{lj}). \tag{15}$$

The sum over j in equation (15) is over nearest neighbours only. Note that G_1 has a single pole of the form

$$\begin{aligned} E(k) &= g\mu_B B_{\text{APP}} + \alpha(S)\langle\hat{T}_0^1\rangle(J(0) + K(0) - J(k)) \\ &= g\mu_B B_{\text{APP}} + \langle S_z\rangle(J(0) + K(0) - J(k)) \end{aligned} \tag{16}$$

in agreement with the usual spin wave result.

Given equations (13) and (16), self-consistent values of $\langle\hat{T}_0^n\rangle$ can be obtained in an iterative manner. In practice, this is achieved by making use of the inverse Fourier transform

$$\langle\langle\hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle = N^{-1}\sum_k G_1 \exp[+\mathrm{i}\,\boldsymbol{k}\cdot(\boldsymbol{R}_l - \boldsymbol{R}_m)] \tag{17}$$

together with the spectral theorem (Zubarev 1960). Proceeding in this fashion, it is easily shown that

$$\langle\hat{T}_{-1}^n\hat{T}_1^1\rangle = a_1[n]\langle\hat{T}_0^n\rangle\varphi \tag{18}$$

where

$$\varphi = \frac{1}{N}\sum_k \frac{1}{[\exp(\beta E_k) - 1]}. \tag{19}$$

Finally, using the contraction properties of the irreducible tensor operators, the left-hand side of equation (18) can be re-expressed in terms of $\langle\hat{T}_0^{n-1}\rangle$, $\langle\hat{T}_0^n\rangle$ and $\langle\hat{T}_0^{n+1}\rangle$ (see Bowden *et al* 1986). Thus equation (18) can be written in the form of a recursion relationship

$$\begin{aligned} &-\left(\frac{(2S-n+1)(2S+n+1)}{(2n-1)(2n+1)}\right)^{1/2}\langle\hat{T}_0^{n-1}\rangle + (1+2\varphi)\langle\hat{T}_0^n\rangle \\ &\qquad + \left(\frac{(2S-n)(2S+n+2)}{(2n+1)(2n+3)}\right)^{1/2}\langle\hat{T}_0^{n+1}\rangle = 0 \end{aligned} \tag{20}$$

which is analogous to Callen and Callen's (1966) generating function for the ensemble averages $\langle S_z^n\rangle$.

3. The $S = 1$ ferromagnet

Since the highest rank tensor that can be supported by a spin-1 system is $n \leqslant 2$, equation (20) can be used to generate two simultaneous equations, for the two unknowns $\langle \hat{T}_0^1 \rangle$ and $\langle \hat{T}_0^2 \rangle$. This leads directly to the solutions

$$\langle \hat{T}_0^1 \rangle = (1 + 2\varphi)/\sqrt{2}(1 + 3\varphi + 3\varphi^2) \tag{21}$$

and

$$\langle \hat{T}_0^2 \rangle = 1/\sqrt{6}[1 + 3\varphi + 3\varphi^2]. \tag{22}$$

Equation (21) was first given by Tahir-Kheli and ter Haar (1962).

In the presence of a crystal field term D, however, it is necessary to obtain a second equation of motion for the Green's function $\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m) \rangle\rangle$ appearing last in equation (12). Proceeding in the usual manner we find, for $S = 1$

$$\begin{aligned} E\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m) \rangle\rangle &= \{a_2[n]\langle \hat{T}_0^{n-1}(m) \rangle + a_3[n]\langle \hat{T}_0^{n+1}(m) \rangle\}\delta_{lm}/(2\pi) \\ &+ g\mu_B B_{APP}\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m) \rangle\rangle + \sqrt{2}\sum_{j \neq l} J_{lj}\{\langle\langle [\hat{T}_1^2(l)\hat{T}_0^1(j) \\ &- \sqrt{3}\hat{T}_0^2(l)\hat{T}_1^1(j) + \sqrt{2}\hat{T}_2^2(l)\hat{T}_{-1}^1(j)]; \hat{T}_{-1}^n(m) \rangle\rangle\} \\ &+ \sqrt{2}\sum_{j \neq l} K_{lj}\langle\langle \hat{T}_1^2(l)\hat{T}_0^1(j); \hat{T}_{-1}^n(m) \rangle\rangle + (\sqrt{3}/\sqrt{2})D\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m) \rangle\rangle \end{aligned} \tag{23}$$

where

$$a_2[n] = -\frac{(n-1)}{2}\left(\frac{(n+3)(3-n)n(n+1)}{(2n-1)(2n+1)}\right)^{1/2} \tag{24}$$

and

$$a_3[n] = -\frac{(n+2)}{2}\left(\frac{(n+4)(2-n)n(n+1)}{(2n+1)(2n+3)}\right)^{1/2}. \tag{25}$$

Once again therefore, it is necessary to decouple the three-operator Green's functions appearing in equation (23).

In the past, a decoupling scheme in the spirit of the RPA has been used:

$$\begin{aligned} &\langle\langle \hat{T}_1^2(l)\hat{T}_0^1(j) - \sqrt{3}\hat{T}_0^2(l)\hat{T}_1^1(j) + \sqrt{2}\hat{T}_2^2(l)\hat{T}_{-1}^1(j)]; \hat{T}_{-1}^n(m) \rangle\rangle \\ &\qquad \simeq \langle \hat{T}_0^1 \rangle\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m) \rangle\rangle - \sqrt{3}\langle \hat{T}_0^2 \rangle\langle\langle \hat{T}_1^1(j); \hat{T}_{-1}^n(m) \rangle\rangle. \end{aligned} \tag{26}$$

Note that $\langle \hat{T}_2^2 \rangle$ has been set equal to zero, because of axial symmetry. Consequently, on substituting equation (26) into (23), we obtain

$$\begin{aligned} E\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m) \rangle\rangle &= \{a_2[n]\langle \hat{T}_0^{n-1}(m) \rangle + a_3[n]\langle \hat{T}_0^{n+1}(m) \rangle\}\delta_{lm}/(2\pi) \\ &+ g\mu_B B_{APP}\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m) \rangle\rangle + \sqrt{2}\sum_{j \neq l}\{[J_{lj} + K_{lj}]\langle \hat{T}_0^1 \rangle\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^1(m) \rangle\rangle \\ &- \sqrt{3}J_{lj}\langle \hat{T}_0^2 \rangle\langle\langle \hat{T}_1^1(j); \hat{T}_{-1}^n(m) \rangle\rangle\} + (\sqrt{3}/\sqrt{2})D\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m) \rangle\rangle. \end{aligned} \tag{27}$$

In practice, equations (12) and equation (27) form a pair of coupled equations, which can be solved for the two Green's functions $\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m) \rangle\rangle$ and $\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m) \rangle\rangle$. However, it should be noted that the terms (diagrams), which have been thrown away in the RPA of equation (11) are most unlikely to be equal to those thrown away in equation (26).

To obtain expressions for the ensemble averages $\langle \hat{T}_0^n \rangle$, it is first necessary to transform both equations (12) and (27). We find

$$E(k)G_1 = a_1[n]\langle \hat{T}_0^n\rangle/(2\pi) + [g\mu_B B_{APP} + \sqrt{2}\langle \hat{T}_0^1\rangle(J(0)+K(0)-J(k))] \times G_1 + (\sqrt{3}/\sqrt{2})DG_2 \tag{28}$$

and

$$E(k)G_2 = \{a_2[n]\langle \hat{T}_0^{n-1}\rangle + a_3[n]\langle \hat{T}_0^{n+1}\rangle\}/(2\pi) + [g\mu_B B_{APP} + \sqrt{2}\langle \hat{T}_0^1\rangle(J(0) + K(0))]G_2 + (\sqrt{3}/\sqrt{2})\{D - 2\langle \hat{T}_0^2\rangle J(k)\}G_1 \tag{29}$$

where (i) G_1 and $J(k)$ have already been defined in equations (14) and (15) respectively, and (ii)

$$G_2 = \sum_m \langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle \exp[-\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_l - \boldsymbol{R}_m)]. \tag{30}$$

Thus the Green's functions G_1 and G_2 are now characterized by the two poles

$$E_{1,2}(k) = g\mu_B B_{APP} + \sqrt{2}\langle \hat{T}_0^1\rangle(J(0) + K(0) - \tfrac{1}{2}J(k)) \pm \tfrac{1}{2}[(\sqrt{2}\langle \hat{T}_0^1\rangle J(k))^2 - 12D\langle \hat{T}_0^2\rangle J(k) + 6D^2]^{1/2}. \tag{31}$$

In practice, equations (28) and (29) must be solved simultaneously to yield explicit solutions for G_1 and G_2. Once these have been determined, expressions for $\langle\langle T_1^1(l), \hat{T}_{-1}^n(m)\rangle\rangle$ and $\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle$ can be obtained via the inverse spatial Fourier transforms

$$\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle = N^{-1}\sum_k G_1 \exp[+\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_l - \boldsymbol{R}_m)] \tag{32}$$

$$\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle = N^{-1}\sum_k G_2 \exp[+\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_l - \boldsymbol{R}_m)]. \tag{33}$$

Further, on making use of the spectral theorem (Zubarev 1960), this time applied to equations (32) and (33), we find

$$\langle \hat{T}_{-1}^n \hat{T}_1^1\rangle = a_1[n]\langle \hat{T}_0^n\rangle\varphi_1 + \{a_2[n]\langle \hat{T}_0^{n-1}\rangle + a_3[n]\langle \hat{T}_0^{n+1}\rangle\}\varphi_2 \tag{34}$$

and

$$\langle \hat{T}_{-1}^n \hat{T}_1^2\rangle = a_1[n]\langle \hat{T}_0^n\rangle\varphi_3 + \{a_2[n]\langle \hat{T}_0^{n-1}\rangle + a_3[n]\langle \hat{T}_0^{n+1}\rangle\}\varphi_4 \tag{35}$$

where (i) $a_1[n]$, $a_2[n]$ and $a_3[n]$ are defined in equations (10), (24) and (25) respectively, and (ii) $\varphi_1 - \varphi_4$ are given by

$$\varphi_1 = \frac{1}{N}\sum_k \frac{1}{E_1 - E_2}\left(\frac{E_1 - E_0}{\mathrm{e}^{\beta E_1} - 1} - \frac{E_2 - E_0}{\mathrm{e}^{\beta E_2} - 1}\right) \tag{36}$$

$$\varphi_2 = \frac{1}{N}\sum_k \frac{\sqrt{3}D}{\sqrt{2}(E_1 - E_2)}\left(\frac{1}{\mathrm{e}^{\beta E_1} - 1} - \frac{1}{\mathrm{e}^{\beta E_2} - 1}\right) \tag{37}$$

$$\varphi_3 = \varphi_2 + \frac{1}{N}\sum_k \frac{-\sqrt{3}\langle \hat{T}_0^2\rangle J(k)}{\sqrt{2}(E_1 - E_2)}\left(\frac{1}{\mathrm{e}^{\beta E_1} - 1} - \frac{1}{\mathrm{e}^{\beta E_2} - 1}\right) \tag{38}$$

$$\varphi_4 = \varphi_1 + \frac{1}{N}\sum_k \frac{-\langle \hat{T}_0^1\rangle J(k)}{\sqrt{2}(E_1 - E_2)}\left(\frac{1}{e^{\beta E_1}-1} - \frac{1}{e^{\beta E_2}-1}\right) \tag{39}$$

respectively, where (i) E_1 and E_2 are given by equation (31), and (ii)

$$E_0 = g\mu_B B_{\mathrm{APP}} + \sqrt{2}\langle \hat{T}_0^1\rangle(J(0) + K(0)). \tag{40}$$

Finally, the contraction properties of irreducible tensor operators can be used to generate the set of ensemble averages for $S = 1$. From equation (34) we find

$$\langle \hat{T}_0^1\rangle = \frac{1 + 2\varphi_1}{\sqrt{2}(1 + 3\varphi_1 + 3\varphi_1^2 + \varphi_2 - 3\varphi_2^2)} \tag{41}$$

and

$$\langle \hat{T}_0^2\rangle = \frac{1 - 2\varphi_2}{\sqrt{6}(1 + 3\varphi_1 + 3\varphi_1^2 + \varphi_2 - 3\varphi_2^2)} \tag{42}$$

whereas from equation (35),

$$\langle \hat{T}_0^1\rangle = \frac{1 + 2\varphi_4}{\sqrt{2}(1 + 3\varphi_4 + 3\varphi_4^2 + \varphi_3 - 3\varphi_3^2)} \tag{43}$$

and

$$\langle \hat{T}_0^2\rangle = \frac{1 - 2\varphi_3}{\sqrt{6}(1 + 3\varphi_4 + 3\varphi_4^2 + \varphi_3 - 3\varphi_3^2)}. \tag{44}$$

Since $\varphi_1 \neq \varphi_4$ and $\varphi_2 \neq \varphi_3$, the ensemble averages calculated from the two Green's functions $\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle$ and $\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle$ are therefore not identical. In fact this problem also exists when $D = 0$. Equations (43) and (44) cannot be used to reproduce the Tahir-Kheli and ter Haar results presented earlier (see equations (21) and (22)).

Many attempts have been made to overcome this problem. Interested readers are referred to the appendix of Devlin (1971), section (B) of Egami and Brooks (1975) and Haley (1978). In particular, Haley has proposed an 'interlevel Green's function', which is essentially a linear addition of the Green's functions involved, with coefficients chosen to ensure global self-consistency. This solution, however, is somewhat artificial in that it does not address the central issue: how can we ensure that ensemble averages calculated using Green's functions $\langle\langle A; B\rangle\rangle$ are independent of the choice of the two operators A and B? A fresh attempt at solving this problem is presented in the following sections.

4. The interaction representation

In nuclear quadrupole resonance (NQR) problems, it is often advantageous to make use of the 'interaction regime' to simplify the description of the nuclear spins, evolving under the influence of RF fields. Consider, for example, the simple case of an $I = 1$ nucleus subject to an axially symmetric quadrupole interaction $\mathcal{H}_Q$, and a time-dependent RF field directed along the x axis. The Hamiltonian for this system can be written in the form

$$\begin{aligned}\mathcal{H} &= \hbar\omega_1 \boldsymbol{I}_x \cos(\omega_Q t) + \mathcal{H}_Q \\ &= -\hbar\omega_1 \hat{T}_1^1(a)\cos(\omega_Q t) + \sqrt{(2/3)}\hbar\omega_Q \hat{T}_0^2\end{aligned} \tag{45}$$

where (i) the $\hat{T}^n_q(\alpha)$ now refer to nuclear operators, (ii) ω_1 is the strength of the RF field directed along the x axis, (iii) ω_Q is the resonant quadrupole frequency.

In many NQR experiments, the quadrupole frequency $\omega_Q \gg \omega_1$, which suggests therefore that it would be advantageous to transform the problem into the 'interaction picture', using the time-dependent unitary transformation

$$\check{U}(t) = \exp[-\mathrm{i}(\mathcal{H}_Q/\hbar)t]. \tag{46}$$

Explicitly

$$\begin{aligned}\mathcal{H}_{\text{int}}/\hbar &= \check{U}(t)^{\dagger}(\mathcal{H}/\hbar)\check{U}(t) - \mathrm{i}\check{U}(t)^{\dagger}\partial\check{U}(t)/\partial t \\ &= -\omega_1\cos(\omega_Q t)\check{U}(t)^{\dagger}\hat{T}^1_1(a)\check{U}(t) \\ &= -\omega_1\{\hat{T}^1_1(a)[1+\cos(2\omega_Q t)] + \mathrm{i}\hat{T}^2_1(s)\sin(2\omega_Q t)\}/2.\end{aligned} \tag{47}$$

In the interaction regime therefore, the strong quadrupole interaction has been 'transformed away', allowing the effect of weaker terms in the Hamiltonian to be examined in more detail.

Following Slichter (1967), terms oscillating rapidly at $2\omega_Q$, in the interaction representation, are dropped. Thus equation (47) simplifies to

$$\mathcal{H}'_{\text{int}}/\hbar = -\frac{\omega_1}{2}\hat{T}^1_1(a) \tag{48}$$

which is time independent. From the NQR point of view, this represents a considerable mathematical simplification. In this approximation, the time dependence of the nuclear density matrix, in the interaction regime, can be expressed in the closed form

$$\rho_{\text{int}}(t) = \exp[-\mathrm{i}(\mathcal{H}'_{\text{int}}/\hbar)t]\rho_{\text{int}}(0)\exp[\mathrm{i}(\mathcal{H}'_{\text{int}}/\hbar)t] \tag{49}$$

which usually forms the starting point for a theoretical discussion of the NQR problem. Interested readers are referred either to Slichter (1967), or Reddy (1988) who has recently discussed NQR for $I = 1$ spin systems in terms of irreducible tensor operators. In the next section however, we return to the problem of the Heisenberg ferromagnet.

5. T$\mathcal{H}$/RPA: the quadrupole interaction representation

First, we shall assume that the quadrupole crystal field $D\hat{T}^2_0$ is strong. This suggests therefore it might be advantageous to 'transform the crystal field terms' away, using the unitary transformation

$$\check{U}(t) = \prod_{j=1}^{N}\exp[\mathrm{i}(D\hat{T}^2_0(j)/\hbar)t] = \exp\left(\mathrm{i}\sum_j(D\hat{T}^2_0(j)/\hbar)t\right) \tag{50}$$

For the Hamiltonian of equation (2) we find (for $S = 1$)

$$\begin{aligned}\mathcal{H}_{\text{int}} &= \check{U}(t)^{\dagger}\mathcal{H}\check{U}(t) - \mathrm{i}\hbar\check{U}(t)^{\dagger}\partial\check{U}(t)/\partial t \\ &= -\sqrt{2}g\mu_{\mathrm{B}}B_{\text{APP}}\left\{\sum_i\hat{T}^1_0(i)\right\} - \sum_{\{i,j\}}(J_{ij}+K_{ij})\hat{T}^1_0(i)\hat{T}^1_0(j) \\ &\quad + \frac{1}{2}\sum_{\{i,j\}}J_{ij}\{(\hat{T}^1_1(i)\hat{T}^1_{-1}(j) + \hat{T}^1_{-1}(i)\hat{T}^1_1(j))^{1/2}[1+\cos(\sqrt{6}Dt/\hbar)]\end{aligned}$$

$$+ (\hat{T}^2_1(i)\hat{T}^2_{-1}(j) + \hat{T}^2_{-1}(i)\hat{T}^2_1(j))^{1/2}(1 - \cos(\sqrt{6}Dt/\hbar))$$
$$+ \mathrm{i}(\hat{T}^2_1(i)\hat{T}^1_{-1}(j) + \hat{T}^1_{-1}(i)\hat{T}^2_1(j) - \hat{T}^1_1(i)\hat{T}^2_{-1}(j)$$
$$- \hat{T}^2_{-1}(i)\hat{T}^1_1(j))^{1/2}\sin(\sqrt{6}Dt/\hbar)\}. \qquad (51)$$

In arriving at equation (51) we have made use of

$$\exp[-\mathrm{i}(D\hat{T}^2_0(j)/\hbar)t]\hat{T}^1_{\pm1}(i)\exp[\mathrm{i}(D\hat{T}^2_0(j)/\hbar)t]$$
$$= \hat{T}^1_{\pm1}(j)\cos(\sqrt{3}Dt/\sqrt{2}\hbar) \mp i\,\hat{T}^2_{\pm1}(j)\sin(\sqrt{3}Dt/\sqrt{2}\hbar) \qquad i = j$$
$$= \hat{T}^1_{\pm1}(i) \qquad i \neq j \qquad (52)$$

and

$$\exp[-\mathrm{i}(D\hat{T}^2_0(j)/\hbar)t]\hat{T}^2_{\pm1}(i)\exp[\mathrm{i}(D\hat{T}^2_0(j)/\hbar)t]$$
$$= \hat{T}^2_{\pm1}(j)\cos(\sqrt{3}Dt/\sqrt{2}\hbar) \mp i\,\hat{T}^1_{\pm1}(j)\sin(\sqrt{3}Dt/\sqrt{2}\hbar) \qquad i = j$$
$$= \hat{T}^2_{\pm1}(i) \qquad i \neq j \qquad (53)$$

which hold for $S = 1$ (note $\gamma(1) = 1$). Thus if we drop the terms oscillating at the high angular frequency of $\sqrt{6}D/\hbar$ in equation (51), we find

$$\mathcal{H}'_{\mathrm{int}} = -\sqrt{2}g\mu_{\mathrm{B}}B_{\mathrm{APP}}\left(\sum_i \hat{T}^1_0(i)\right) - \sum_{\{i,j\}}(J_{ij} + K_{ij})\hat{T}^1_0(i)\hat{T}^1_0(j)$$
$$+ \tfrac{1}{2}\sum_{\{i,j\}} J_{ij}\,(\hat{T}^1_1(i)\hat{T}^1_{-1}(j) + \hat{T}^1_{-1}(i)\hat{T}^1_1(j)$$
$$+ \hat{T}^2_1(i)\hat{T}^2_{-1}(j) + \hat{T}^2_{-1}(i)\hat{T}^2_1(j)). \qquad (54)$$

Note (i) the appearance of the rank 2 tensors in the exchange interaction terms of equation (54), and (ii) the truncation is really only valid when the crystal field parameter D is strong. The latter point is taken up again in section 6.

Now that the important exchange terms have been identified, it is instructive to transform back to the laboratory frame. Rather surprisingly, it is found that equation (54) is 'invariant' with respect to the inverse quadrupolar transformation of equation (48). Thus in the laboratory frame the truncated Hamiltonian is given by

$$\mathcal{H}_{\mathrm{mod}} = \mathcal{H}'_{\mathrm{int}} + \mathcal{H}_{\mathrm{D}} \qquad (55)$$

where we have now recovered the full crystal field Hamiltonian $\mathcal{H}_{\mathrm{D}}$.

We are now in a position to implement the RPA. This yields two coupled Green's function equations of motion, for the $S = 1$ ferromagnet. Explicitly

$$E\langle\langle\hat{T}^1_1(l);\hat{T}^n_{-1}(m)\rangle\rangle = a_1[n]\langle\hat{T}^n_0(m)\rangle\delta_{lm}/(2\pi) + g\mu_{\mathrm{B}}B_{\mathrm{APP}}\langle\langle\hat{T}^1_1(l);\hat{T}^n_{-1}(m)\rangle\rangle$$
$$+ \sqrt{2}\sum_{j\neq l}[(J_{lj} + K_{lj})\langle\langle\hat{T}^1_1(l)\hat{T}^1_0(j);\hat{T}^n_{-1}(m)\rangle\rangle - \tfrac{1}{2}J_{lj}\langle\langle\hat{T}^1_0(l)\hat{T}^1_1(j);\hat{T}^n_{-1}(m)\rangle\rangle]$$
$$- \frac{1}{2}\sum_{j\neq l} J_{lj}[\sqrt{6}\langle\langle\hat{T}^2_0(l)\hat{T}^2_1(j);\hat{T}^n_{-1}(m)\rangle\rangle + 2\langle\langle\hat{T}^2_2(l)\hat{T}^2_{-1}(j);\hat{T}^n_{-1}(m)\rangle\rangle]$$
$$+ (\sqrt{3}/\sqrt{2})D\langle\langle\hat{T}^2_1(l);\hat{T}^n_{-1}(m)\rangle\rangle \qquad (56)$$

and

$$E\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle = (a_2[n]\langle \hat{T}_0^{n-1}(m)\rangle + a_3[n]\langle \hat{T}_0^{n+1}(m)\rangle)\delta_{lm}/(2\pi)$$
$$+ g\mu_B B_{APP}\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle + \sum_{j\neq l}[\sqrt{2}(J_{lj}+K_{lj})$$
$$\times \langle\langle \hat{T}_1^2(l)\hat{T}_0^1(j); \hat{T}_{-1}^n(m)\rangle\rangle + J_{lj}\langle\langle \hat{T}_2^2(l)\hat{T}_{-1}^1(j); \hat{T}_{-1}^n(m)\rangle\rangle]$$
$$-\frac{1}{2}\sum_{j\neq l} J_{lj}(\sqrt{6}\langle\langle \hat{T}_0^2(l)\hat{T}_1^1(j); \hat{T}_{-1}^n(m)\rangle\rangle + \sqrt{2}\langle\langle \hat{T}_0^1(l)\hat{T}_1^2(j); \hat{T}_{-1}^n(m)\rangle\rangle)$$
$$+ (\sqrt{3}/\sqrt{2})D\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle. \qquad (57)$$

On making the RPA therefore, and invoking translational invariance, we obtain the two equations

$$E\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle = a_1[n]\langle \hat{T}_0^n(m)\rangle\delta_{lm}/(2\pi) + g\mu_B B_{APP}\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle$$
$$+\sqrt{2}\sum_{j\neq l}\langle \hat{T}_0^1\rangle[(J_{lj}+K_{lj})\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle - \tfrac{1}{2}J_{lj}\langle\langle \hat{T}_1^1(j); \hat{T}_{-1}^n(m)\rangle\rangle]$$
$$-\frac{1}{2}\sum_{j\neq l} J_{lj}\sqrt{6}\langle \hat{T}_0^2\rangle\langle\langle \hat{T}_1^2(j); \hat{T}_{-1}^n(m)\rangle\rangle + (\sqrt{3}/\sqrt{2})D\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle \qquad (58)$$

and

$$E\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle = (a_2[n]\langle \hat{T}_0^{n-1}(m)\rangle + a_3[n]\langle \hat{T}_0^{n-1}(m)\rangle)\delta_{lm}/(2\pi)$$
$$+ g\mu_B B_{APP}\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle + \sum_{j\neq 1}\langle \hat{T}_0^1\rangle\sqrt{2}(J_{lj}+K_{lj})\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle$$
$$-\frac{1}{2}\sum_{j\neq l} J_{lj}(\sqrt{6}\langle \hat{T}_0^2\rangle\langle\langle \hat{T}_1^1(j); \hat{T}_{-1}^n(m)\rangle\rangle + \sqrt{2}\langle \hat{T}_0^1\rangle\langle\langle \hat{T}_1^2(j); \hat{T}_{-1}^n(m)\rangle\rangle)$$
$$+ (\sqrt{3}/\sqrt{2})D\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle. \qquad (59)$$

Consequently, on taking the spatial Fourier transforms,

$$E(k)G_1 = a_1[n]\langle \hat{T}_0^n\rangle/(2\pi) + [g\mu_B B_{APP} + \sqrt{2}\langle \hat{T}_0^1\rangle(J(0)+K(0)-\tfrac{1}{2}J(k))]G_1$$
$$+ (\sqrt{3}/\sqrt{2})[D - \langle \hat{T}_0^2\rangle J(k)]G_2 \qquad (60)$$

$$E(k)G_2 = (a_2[n]\langle \hat{T}_0^{n-1}\rangle + a_3[n]\langle \hat{T}_0^{n+1}\rangle)/(2\pi) + (\sqrt{3}/\sqrt{2})[D - \langle \hat{T}_0^2\rangle J(k)]G_1$$
$$+ [g\mu_B B_{APP} + \sqrt{2}\langle \hat{T}_0^1\rangle(J(0)+K(0)-\tfrac{1}{2}J(k))]G_2 \qquad (61)$$

where G_1, G_2 and $J(k)$ have been defined in equations (14), (30) and (15), respectively. Thus the two poles of both the Green's functions G_1 and G_2 are given by

$$E_{1,2}(k) = [g\mu_B B_{APP} + \sqrt{2}\langle \hat{T}_0^1\rangle(J(0)+K(0)-\tfrac{1}{2}J(k)) \pm (\sqrt{3}/\sqrt{2})(D - \langle \hat{T}_0^2\rangle J(k)). \qquad (62)$$

Note that the excitation energies are characterized by (i) crystal field excitons in the limit $D\to\infty$, and (ii) spin waves in the limit $D\to 0$.

Equations (60) and (61) bear some resemblance to that of the previous RPA result embodied in equations (28) and (29). However, in the transformed Hamiltonian random phase approximation (T𝒦/RPA) approach the two equilibrium correlation functions are of the form

$$\langle \hat{T}^{n}_{-1}\hat{T}^{1}_{1}\rangle = a_1[n]\langle \hat{T}^{n}_{0}\rangle\varphi_5 + (a_2[n]\langle \hat{T}^{n-1}_{0}\rangle + a_3[n]\langle \hat{T}^{n+1}_{0}\rangle)\varphi_6 \tag{63}$$

and

$$\langle \hat{T}^{n}_{-1}\hat{T}^{2}_{1}\rangle = a_1[n]\langle \hat{T}^{n}_{0}\rangle\varphi_6 + (a_2[n]\langle \hat{T}^{n-1}_{0}\rangle + a_3[n]\langle \hat{T}^{n+1}_{0}\rangle)\varphi_5 \tag{64}$$

where

$$\varphi_5 = \frac{1}{N}\sum_k \frac{1}{2}\left(\frac{1}{e^{\beta E_1} - 1} + \frac{1}{e^{\beta E_2} - 1}\right) \tag{65}$$

$$\varphi_6 = \frac{1}{N}\sum_k \frac{1}{2}\left(\frac{1}{e^{\beta E_1} - 1} - \frac{1}{e^{\beta E_2} - 1}\right). \tag{66}$$

Again two equations (63) and (64), can be used independently to generate the set of correlation functions for $S = 1$. However due to the symmetry of equations (63) and (64), the unique solutions

$$\langle \hat{T}^{1}_{0}\rangle = \frac{1 + 2\varphi_5}{\sqrt{2}(1 + 3\varphi_5 + 3\varphi_5^2 + \varphi_6 - 3\varphi_6^2)} \tag{67}$$

and

$$\langle \hat{T}^{2}_{0}\rangle = \frac{1 - 2\varphi_6}{\sqrt{6}(1 + 3\varphi_5 + 3\varphi_5^2 + \varphi_6 - 3\varphi_6^2)} \tag{68}$$

are obtained, irrespective of the size of D. The underlying reason for this behaviour can be traced to the symmetry of the coupling coefficients in the Green's function equations (60) and (61).

In the next section, we show that equations (62), (64) and (65) would appear to hold for lower values of D, than might be anticipated at first sight.

6. Energy gap considerations

In this, and the following section, we try to set limits on the regions where the T𝒦/RPA model might be expected to hold.

In the first place, we set the crystal field parameter $D \equiv 0$. The two excitation branches of equation (62) are given by

$$E_1(k)_{D=0} = g\mu_B B_{APP} + \sqrt{2}\langle \hat{T}^{1}_{0}\rangle(J(0) + K(0)) - (1/\sqrt{2})(\langle \hat{T}^{1}_{0}\rangle + \sqrt{3}\langle \hat{T}^{2}_{0}\rangle)J(k) \tag{69}$$

$$E_2(k)_{D=0} = g\mu_B B_{APP} + \sqrt{2}\langle \hat{T}^{1}_{0}\rangle(J(0) + K(0)) - (1/\sqrt{2})(\langle \hat{T}^{1}_{0}\rangle - \sqrt{3}\langle \hat{T}^{2}_{0}\rangle)J(k) \tag{70}$$

which can be compared to the well known Tahir-Kheli and ter Haar result of equation

(15). In the limit $T = 0$ K, $\langle \hat{T}_0^1 \rangle = \surd 3 \langle \hat{T}_0^2 \rangle$. Thus the two excitation branches reduce to a dispersive branch

$$E_1(k)_{\substack{D=0\\T=0\,\mathrm{K}}} = g\mu_B B_{APP} + \surd 2 \langle \hat{T}_0^1 \rangle (J(0) + K(0) - J(k)) \tag{71}$$

and a non-dispersive branch

$$E_2(k)_{\substack{D=0\\T=0\,\mathrm{K}}} = g\mu_B B_{APP} + \surd 2 \langle \hat{T}_0^1 \rangle (J(0) + K(0)). \tag{72}$$

The first of these can be identified with the usual spin wave result. At absolute zero, only the $|I_z = 1\rangle$ ground state is populated, and so collective modes result from excitations involving transitions from ground state $|I_z = 1\rangle$ to first excited state $|I_z = 0\rangle$. The non-dispersive branch $E_2(k)$, on the other hand, corresponds to a single-ion excitation from $|I_z = 0\rangle$ to $|I_z = -1\rangle$, which at $T = 0$ K can only involve non-populated states. However, as the temperature is raised, $\langle \hat{T}_0^2 \rangle$ will differ from its saturated value, and two dispersive branches will emerge.

From an examination of equation (62), it is evident that the energy gap $\Delta(E_1(0))$, can be written in the form

$$\Delta = (\surd 3/\surd 2)D + g\mu_B B_{APP} + \surd 2 \langle \hat{T}_0^1 \rangle K(0) + (1/\surd 2)J(0)(\langle \hat{T}_0^1 \rangle - \surd 3 \langle \hat{T}_0^2 \rangle). \tag{73}$$

As noted earlier, the last term in equation (73) vanishes at $T = 0$ K because $\langle \hat{T}_0^1 \rangle \equiv \surd 3 \langle \hat{T}_0^2 \rangle$ at saturation. However, at higher temperatures it is possible, in the limit $D \to 0$, that $\langle \hat{T}_0^2 \rangle$ will fall more rapidly that $\langle \hat{T}_0^1 \rangle$ thereby giving rise to an unphysical energy gap which increases with increasing temperature. By way of contrast the presence of a large crystal field parameter D ensures that $\langle \hat{T}_0^1 \rangle$ will decrease with increasing temperature more quickly than $\langle \hat{T}_0^2 \rangle$. Indeed at the Curie temperature, $\langle \hat{T}_0^2 \rangle$ is necessarily finite while $\langle \hat{T}_0^1 \rangle$ is identically zero. Thus in this case the energy gap Δ will decrease with increasing temperature.

To probe this question further, self-consistent calculations have been carried out for small values of D and $K(0)$. From these results it is possible to conclude that the T$\mathcal{H}$/RPA model predicts physically sensible results when $D/J(0) \geq \surd(2/3)$, and/or $K(0)/J(0) \geq 1$. This question is taken up again in the next section, where various estimates of the Curie temperature are compared and discussed.

7. The Curie temperature

In this section we briefly compare the Curie temperature T_C obtained using the T$\mathcal{H}$/RPA method with those obtained by other authors.

In the first place, we set the crystal field parameter D equal to zero. In this case it is easily shown, using equation (34), that the Curie temperature T_C is given by

$$k_B T_C = \frac{2}{3}\left(\frac{1}{N}\sum_k \frac{1}{J(0) + K(0) - J(k)}\right)^{-1} \tag{74}$$

which is the usual Tahir-Kheli and ter Haar result for $S = 1$. However, if we use equation (35), we find

$$k_B T_C = \frac{2}{3}\left(\frac{1}{N}\sum_k \frac{1}{J(0) + K(0)}\right)^{-1} \tag{75}$$

which is the molecular field result. It is clear therefore that the RPA results are not independent of the choice of Green's function, as noted earlier in section 3.

From the T$\mathscr{H}$/RPA results presented in this paper we obtain the unique solution

$$k_B T_C = \frac{2}{3}\left(\frac{1}{N}\sum_k \frac{1}{J(0)+K(0)-\frac{1}{2}J(k)}\right)^{-1} \tag{76}$$

which can be compared to equations (74) and (75). As noted in the previous section, the T$\mathscr{H}$/RPA solution only gives physically reasonable results at finite temperatures when $D=0$, provided $K(0)/J(0) \geqslant 1$. A comparison of the calculated Curie temperatures obtained using equations (74) (RPA_1), (75) (RPA_2) and (76) (T$\mathscr{H}$/RPA) for various ratios of $K(0)/J(0)$ can be seen in figure 1. In the limit $K(0)\to\infty$, all the three estimates for T_C converge.

The situation for $D>0$ is more complex because $\langle\hat{T}_0^2\rangle$ is now finite at T_C. In this case it can be shown, using the non-consistent RPA solution (RPA_1), based on equation (34), that the Curie temperature T_C is given by

$$k_B T_C = \frac{\sqrt{3}\langle\hat{T}_0^2\rangle}{(1-2\theta_1)}\left(\frac{2}{N}\sum_k \frac{\sqrt{2}[J(0)+K(0)-\frac{1}{2}J(k)]}{2\{1-\cosh[\beta\frac{1}{2}(6D^2-12D\langle\hat{T}_0^2\rangle J(k))^{1/2}]\}}\right) \tag{77}$$

where (i)

$$\langle\hat{T}_0^2\rangle = \frac{2\sqrt{2}}{\sqrt{3}}\frac{1}{(1+6\sigma_1)} \tag{78}$$

and (ii)

$$\theta_1 = -\frac{1}{N}\sum_k \frac{\sqrt{6}D-\sqrt{6}\langle\hat{T}_0^2\rangle J(k)}{2(6D^2-12D\langle\hat{T}_0^2\rangle J(k))^{1/2}}\left(\frac{\sinh[\beta\frac{1}{2}(6D^2-12D\langle\hat{T}_0^2\rangle J(k))^{1/2}]}{1-\cosh[\beta\frac{1}{2}(6D^2-12D\langle\hat{T}_0^2\rangle J(k))^{1/2}]}\right) \tag{79}$$

and (iii)

$$\sigma_1 = -\frac{1}{N}\sum_k \frac{\sqrt{6}D}{2(6D^2-12D\langle\hat{T}_0^2\rangle J(k))^{1/2}}\left(\frac{\sinh[\beta\frac{1}{2}(6D^2-12D\langle\hat{T}_0^2\rangle J(k))^{1/2}]}{1-\cosh[\beta\frac{1}{2}(6D^2-12D\langle\hat{T}_0^2\rangle J(k))^{1/2}]}\right). \tag{80}$$

Alternatively if we use the other non-consistent RPA solution (RPA_2), based on equation (35), we find

$$k_B T_C = \frac{\sqrt{3}\langle\hat{T}_0^2\rangle}{(1-2\theta_1)}\left(\frac{2}{N}\sum_k \frac{\sqrt{2}(J(0)+K(0)-\frac{1}{2}J(k))}{2\{1-\cosh[\beta\frac{1}{2}(6D^2-12D\langle\hat{T}_0^2\rangle J(k))^{1/2}]\}}\right) \tag{81}$$

where (i)

$$\langle\hat{T}_0^2\rangle = \frac{2\sqrt{2}}{\sqrt{3}}\frac{1}{(1+6\sigma_2)} \tag{82}$$

(ii) θ_1 is given above in equation (79),

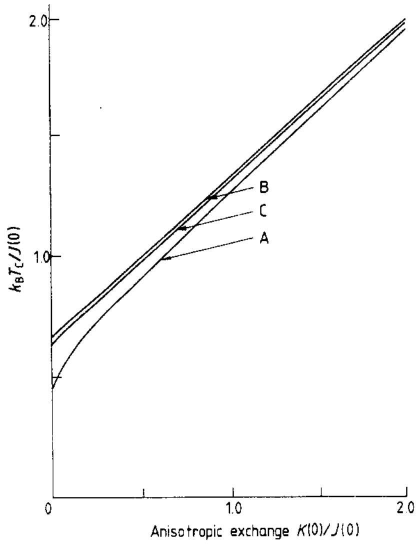

Figure 1. The Curie temperature as a function of the anisotropic exchange $K(0)$, for $D = 0$, as given by A, the RPA$_1$ (equation (74)), B, the RPA$_2$ (equation (75)) and C, the T$\mathcal{H}$/RPA (equation (76)). These results have been obtained using a simple cubic lattice, which of course cannot support an axial crystal field. Nevertheless we have chosen to use this lattice for comparison purposes.

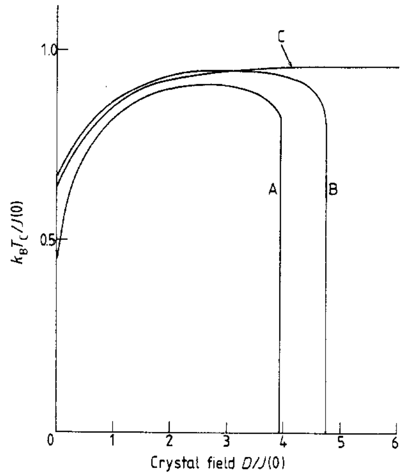

Figure 2. The Curie temperature as a function of the crystal field parameter D, for $K(0) = 0$, as given by A, the RPA$_1$ (equation (77)), B, the RPA$_2$ (equation (81)) and C, the T$\mathcal{H}$/RPA (equation (84)).

and (iii)

$$\sigma_2 = -\frac{1}{N}\sum_k \frac{\surd 6D - 2\surd 6\langle \hat{T}_0^2\rangle J(k)}{2(6D^2 - 12D\langle \hat{T}_0^2\rangle J(k))^{1/2}}\left(\frac{\sinh[\beta\frac{1}{2}(6D^2 - 12D\langle \hat{T}_0^2\rangle J(k))^{1/2}]}{1-\cosh[\beta\frac{1}{2}(6D^2 - 12D\langle \hat{T}_0^2\rangle J(k))^{1/2}]}\right). \tag{83}$$

Finally, the T$\mathcal{H}$/RPA model, based on equations (63) and (64), predicts that

$$k_B T_C = \frac{\surd 3\langle \hat{T}_0^2\rangle}{(1-2\theta_3)}\left(\frac{2}{N}\sum_k \frac{\surd 2(J(0) + K(0) - \frac{1}{2}J(k))}{2\{1-\cosh[\beta(\surd 3/\surd 2)(D-\langle \hat{T}_0^2\rangle J(k))]\}}\right) \tag{84}$$

where (i)

$$\langle \hat{T}_0^2\rangle = \frac{2\surd 2}{\surd 3}\frac{1}{(1+6\theta_3)} \tag{85}$$

and (ii)

$$\theta_3 = -\frac{1}{N}\sum_k \frac{1}{2}\left(\frac{\sinh[\beta(\surd 3/\surd 2)(D-\langle \hat{T}_0^2\rangle J(k))]}{1-\cosh[\beta(\surd 3/\surd 2)(D-\langle \hat{T}_0^2\rangle J(k))]}\right). \tag{86}$$

As noted earlier, the T$\mathcal{H}$/RPA result is only physically reasonable at finite temperatures, when $D/J(0) \geqslant \surd(2/3)$.

Since $\langle \hat{T}_0^2\rangle$ is temperature dependent in all three models presented above, it is necessary to solve for both $\langle \hat{T}_0^2\rangle$ and k_BT_C simultaneously. In figure 2, the computed

Table 1. Predicted Curie temperatures, $k_B T_C/J(0)$

(*a*) $D \to \infty$ ($K(0) = 0$)

Lattice	Series (Ising doublet) Fisher (1967)	T$\mathscr{H}$/RPA	Mean field theory	RPA$_1$	RPA$_2$
SC	0.7517	0.9563	1.0	0.0	0.0
FCC	0.8162	0.9687	1.0	0.0	0.0

(*b*) $D \to 0$ ($K(0) = 0$)

Lattice	Series (Heisenberg $S = 1$) Fisher (1967)	T$\mathscr{H}$/RPA	Mean field theory	RPA$_1$	RPA$_2$
SC	0.440	0.6368	0.666	0.440	0.666
FCC	0.498	0.6468	0.666	0.49	0.666

Note (i) the high-temperature-series results for $D \to 0$ have been scaled by a factor of $\frac{2}{3}$ to compare with our results, and (ii) the RPA$_1$ results for $D \to 0$ are the well known Tahir-Kheli and ter Haar values which agree remarkably well with the high-temperature-series results.

values of T_C (for $K(0) = 0$) in the RPA$_1$, RPA$_2$ and T$\mathscr{H}$/RPA models, can be seen as a function of $D/J(0)$. From an examination of this data it will be seen that as $D \to \infty$, the predicted value of T_C, obtained using the T$\mathscr{H}$/RPA saturates at $0.9563J(0)$ for a SC lattice. This is not entirely unexpected since molecular field theory predicts a limit of $T_C = J(0)$. As $D \to \infty$, the crystal field ground state is an $|Sz = \pm 1\rangle$ doublet. Consequently, the magnetic exchange terms can only act within the $|\pm 1\rangle$ doublet, with little influence from the high energy $|0\rangle$ singlet state. By way of contrast, it will be seen that the calculated Curie temperature based on the non-consistent RPA models of equations (77) and (81) go to zero above $D/J(0) = 4$ and 5 respectively. This unphysical behaviour was first highlighted by Egami and Brooks (1975, p 1027). In particular, they set their $\varphi_{22} = 0$, in order to obtain physically sensible expressions for T_C in the limit $D \to \infty$.

Finally, in table 1 the predicted Curie temperatures for the T$\mathscr{H}$/RPA model in the two limits $D \to \infty$ and $D \to 0$, are compared with the Green's function results of Tahir-Kheli and ter Haar (1962), Egami and Brooks (1975), Devlin (1971), and the high-temperature-series results (see for example the review by Fisher (1967)). It will be observed that the Curie temperatures of the T$\mathscr{H}$/RPA for small D are disappointingly close to the mean field results. However, in practice it may be possible to achieve better agreement with the high-temperature series by modifying the RPA in the spirit of the Callen (1963) decoupling scheme.

8. Conclusion

A new method of incorporating strong crystal fields into the RPA model of an $S = 1$ ferromagnet has been presented and discussed. In particular, it has been shown that the T$\mathscr{H}$/RPA can be used to obtain a unique set of ensemble averages $\langle \hat{T}_0^n \rangle$, starting with the Green's function equation of motion for either $\langle\langle \hat{T}_{\pm 1}^1(l); \hat{T}_{\mp 1}^n(m) \rangle\rangle$ or

$\langle\langle \hat{T}^2_{\pm 1}(l); \hat{T}^n_{\mp 1}(m)\rangle\rangle$, in contrast to previous work. In addition, it has been shown (i) a unique solution can be obtained for the Curie temperature T_C, regardless of the strength of the crystal field parameter D, and (ii) the usual spin wave result is obtained in the limit $T \to 0$ K and $D \to 0$. In a following paper, it will be shown that the T$\mathscr{H}$/RPA for an $S = 1$ spin system can also be used to obtain a two-parameter analogue of the Callen and Shtrikman (1965) generating function, for the ensemble averages $\langle \hat{T}^n_0 \rangle$.

Finally, it should be stressed that the results obtained in this paper hold specifically for $S = 1$ spin ensembles. For spins $S > 1$, higher rank tensors $\hat{T}^n_q$ with $n > 2$ will be generated when the Heisenberg exchange Hamiltonian is transformed into the interaction representation. Thus the tensor algebra will be more complex. Nevertheless, in another paper it will be demonstrated that (i) the T$\mathscr{H}$/RPA does give rise to a unique set of ensemble averages $\langle \hat{T}^n_0 \rangle$ for $S = \frac{3}{2}$ spin ensembles, and (ii) a two-parameter analogue of the Callen and Shtrikman (1965) generating function for the ensemble averages $\langle \hat{T}^n_0 \rangle$ can be found.

References

Bowden G J, Martin J P D and Oitmaa J 1986 *J. Phys. C: Solid State Phys.* **19** 551–61

Callen H B 1963 *Phys. Rev.* **130** 890–8

Callen H B and Callen E 1966 *J. Phys. Chem. Solids* **27** 1271–85

Callen H B and Shtrikman S 1965 *Solid State Commun.* **3** 5–9

Das T P and Hahn E L 1958 *Solid State Physics, Nuclear Quadrupole Resonance Spectroscopy* (New York: Academic) Suppl. 1

Devlin J F 1971 *Phys. Rev.* B **4** 136–46

Egami T and Brooks M S S 1975 *Phys. Rev.* B **12** 1021–8

Fisher M E 1967 *Reports on Progress in Physics* vol. XXX, part II, 615–730

Haley S B 1978 *Phys. Rev.* B **17** 337–46

Haley S B and Erdös P 1972 *Phys. Rev.* B **5** 1106–19

Slichter C P 1963 *Principles of Magnetic Resonance* (New York: Harper and Row)

Tahir-Kheli and ter Haar D 1962 *Phys. Rev.* **127** 88–94

Tyablikov S V 1959 *Ukr. Mat. Zh.* **11** 287–94

Zubarev D N 1960 *Sov. Phys.–Usp.* **3** 320–45