# The random phase approximation and crystal field effects in magnetism: S=3/2



1990 J. Phys.: Condens. Matter 2 10461

(http://iopscience.iop.org/0953-8984/2/51/018)

J. Phys.: Condens. Matter 2 (1990) 10461–10473. Printed in the UK

# The random phase approximation and crystal field effects in magnetism: $S = 3/2$

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

Received 22 May 1990, in final form 24 September 1990

**Abstract.** A novel solution to the problem of incorporating crystal field interactions into the RPA model of an Heisenberg ferromagnet, known as the transformed Hamiltonian/random phase approximation (TH/RPA), is applied to an easy axis $S = 3/2$ Heisenberg ferromagnet, subject to an axially symmetric quadrupole interaction. It is demonstrated that unique solutions can be obtained for the ensemble averages. In addition, it is shown that (i) in the limit $T \rightarrow 0$ K and $D \rightarrow 0$ the usual spin wave result is obtained, (ii) three excitation branches are present, which exhibit dispersion at finite temperatures, (iii) a unique solution can be obtained for the Curie temperature $T_C$ in the presence of crystal fields, in contrast to earlier work, and (iv) a two-parameter analogue of the Callen and Shtrikman single-particle generating function can be constructed which exactly mimics the TH/RPA ensemble averages.

## 1. Introduction

Incorporating crystal field interactions into the random phase approximation (RPA) for a Heisenberg ferromagnet has been an unresolved problem in the literature for many years (see for example Devlin (1971), Haley and Erdos (1972), Egami and Brooks (1975) and Haley (1978)). The central problem is that of devising a consistent decoupling scheme which allows ensemble averages to be calculated in a unique manner from the differing Green function equations which arise. Recently, however, a novel solution to this problem has been devised by Bowden and Martin (1990a), using the simple example of an easy axis spin $S = 1$ ferromagnet, subject to both Heisenberg exchange and an axially symmetric quadrupole crystal field interaction. The method employed by these authors has been termed the transformed Hamiltonian/random phase approximation (TH/RPA).

In this paper, the T$\mathcal{H}$/RPA is applied to the case of an easy axis $S = 3/2$ ferromagnet. As with the $S = 1$ case, the T$\mathcal{H}$/RPA is found to yield unique solutions for the ensemble averages $\langle \hat{T}_0^1 \rangle$, $\langle \hat{T}_0^2 \rangle$ etc. In addition, it is shown that (i) the three excitation branches (for $S = 3/2$) exhibit dispersion at finite temperatures, (ii) in the limit $T \rightarrow 0$ K and $D \rightarrow 0$, the usual spin wave result is obtained, and (iii) the T$\mathcal{H}$/RPA solution can be exactly mimicked by an equivalent two-parameter effective single-particle model, in the spirit of Callen and Shtrikman (1965).

Finally, both the T$\mathcal{H}$/RPA and RPA are used to calculate the Curie temperature $T_C$ for various values of the crystal field parameter $D$ and the magnetic exchange anisotropy $K(0)$. It is found that the T$\mathcal{H}$/RPA results are disappointingly close to the molecular field predictions. Nevertheless, the T$\mathcal{H}$/RPA represents a considerable advance over earlier modifications of RPA theory which showed unphysical behaviour in that $T_C \to 0$ as $D \to \infty$.

## 2. $S = 3/2$ Heisenberg ferromagnet with an axially symmetric quadratic crystal field

The Hamiltonian in question can be written in the form

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

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

Following Bowden and Martin (1990a), however, we choose to use unit irreducible tensor operators $\hat{T}_q^n$, in place of the Cartesian operators $S_x$ etc, because of their superior commutation, construction, contraction and rotational properties. In tensorial form, for $S = 3/2$, therefore

$$\mathcal{H} = -\sqrt{5} g\mu_B B_{APP}\{\Sigma_i \hat{T}_0^1(i)\} - \tfrac{1}{2}\Sigma_{\{i,j\}} 5J_{ij}[\hat{T}_0^1(i)\hat{T}_0^1(j) - \hat{T}_1^1(i)\hat{T}_{-1}^1(j) - \hat{T}_{-1}^1(i)\hat{T}_1^1(j)]$$
$$- \tfrac{1}{2}\Sigma_{\{i,j\}} 5K_{ij}\hat{T}_0^1(i)\hat{T}_0^1(j) - \sqrt{6} D\{\Sigma_i \hat{T}_0^2(i)\} \tag{2}$$

where the $\hat{T}_q^n(\alpha)$ are defined, for example, by Buckmaster *et al* (1972) and Bowden *et al* (1986).

## 3. TH/RPA: the quadrupole interaction representation

We first make use of the 'interaction representation' to transform away the strong single-ion crystal field terms in equations (1) and (2) (Bowden and Martin 1990a). Using the time-dependent unitary transformation

$$\check{U}(t) = \prod_{j=1}^{N} \exp\{\mathrm{i}[\sqrt{6} D\hat{T}_0^2(j)/\hbar]t\} = \exp\{\mathrm{i}\sum_j [\sqrt{6} D\hat{T}_0^2(j)/\hbar]t\} \tag{3}$$

we find, after some manipulation,

$$\mathcal{H}_{int} = \check{U}(y)^\dagger \mathcal{H}\check{U}(t) - \mathrm{i}\hbar\check{U}(t)^\dagger\,\partial\check{U}(t)/\partial t$$
$$= -\sqrt{5} g\mu_B B_{APP}\{\Sigma_i \hat{T}_0^1(i)\} - \tfrac{5}{2}\sum_{\{i,j\}}(J_{ij} + K_{ij})\hat{T}_0^1(i)\hat{T}_0^1(j)$$
$$+ \tfrac{5}{2}\sum_{\{i,j\}} J_{ij}\{[\hat{T}_0^1(i)\hat{T}_{-1}^1(j) + \hat{T}_{-1}^1(i)\hat{T}_1^1(j)][\tfrac{2}{5} + \tfrac{3}{5}\cos(\sqrt{6}Dt/\hbar)]^2$$
$$+ [\hat{T}_1^2(i)\hat{T}_{-1}^2(j) + \hat{T}_{-1}^2(i)\hat{T}_1^2(j)]\tfrac{3}{5}[\sin(\sqrt{6}Dt/\hbar)]^2$$
$$+ [\hat{T}_1^3(i)\hat{T}_{-1}^3(j) + \hat{T}_{-1}^3(i)\hat{T}_1^3(j)]\tfrac{6}{25}[\cos(\sqrt{6}Dt/\hbar) - 1]^2$$

$$- \mathrm{i}[\hat{T}^2_1(i)\hat{T}^1_{-1}(i)\hat{T}^2_1(j) - \hat{T}^1_1(i)\hat{T}^2_{-1}(j)$$

$$- \hat{T}^2_{-1}(i)\hat{T}^1_1(j)][\tfrac{2}{5} + \tfrac{3}{5}\cos(\sqrt{6}Dt/\hbar)](\sqrt{3}/\sqrt{5})\sin(\sqrt{6}Dt/\hbar)$$

$$+ [\hat{T}^3_1(i)\hat{T}^1_{-1}(j) + \hat{T}^1_{-1}(i)\hat{T}^3_1(j) + \hat{T}^1_1(i)\hat{T}^3_{-1}(j)$$

$$+ \hat{T}^3_{-1}(i)\hat{T}^1_1(j)][\tfrac{2}{5} + \tfrac{3}{5}\cos(\sqrt{6}Dt/\hbar)](\sqrt{6}/5)[\cos(\sqrt{6}Dt/\hbar) - 1]$$

$$- \mathrm{i}[\hat{T}^2_1(i)\hat{T}^3_{-1}(j) + \hat{T}^3_{-1}(i)\hat{T}^2_1(j) - \hat{T}^3_1(i)\hat{T}^2_{-1}(j)$$

$$- \hat{T}^2_{-1}(i)\hat{T}^3_1(j)](\sqrt{6}/5)[\cos(\sqrt{6}Dt/\hbar) - 1]$$

$$\times (\sqrt{3}/\sqrt{5})\sin(\sqrt{6}Dt/\hbar)\} \tag{4}$$

where use has been made of the identities

$$\check{U}(t)^\dagger \hat{T}^1_{\pm1}(i)\check{U}(t) = \hat{T}^1_{\pm1}(i)[\tfrac{2}{5} + \tfrac{3}{5}\cos(\sqrt{6}Dt/\hbar)] \mp \mathrm{i}\hat{T}^2_{\pm1}(i)(\sqrt{3}/\sqrt{5})\sin(\sqrt{6}Dt/\hbar) + \hat{T}^3_{\pm1}(i)(\sqrt{6}/5)[\cos(\sqrt{6}Dt/\hbar) - 1] \tag{5}$$

$$\check{U}(t)^\dagger \hat{T}^2_{\pm1}(i)\check{U}(t) = \mp \mathrm{i}\hat{T}^1_{\pm1}(i)(\sqrt{3}/\sqrt{5})\sin(\sqrt{6}Dt/\hbar) + \hat{T}^2_{\pm1}(i)\cos(\sqrt{6}Dt/\hbar) \mp \mathrm{i}\hat{T}^3_{\pm1}(i)(\sqrt{2}/\sqrt{5})\sin(\sqrt{6}Dt/\hbar) \tag{6}$$

$$\check{U}(t)^\dagger \hat{T}^3_{\pm1}(i)\check{U}(t) = \hat{T}^1_{\pm1}(i)(\sqrt{6}/5)[\cos(\sqrt{6}Dt/\hbar) - 1] \mp \mathrm{i}\hat{T}^2_{\pm1}(i)(\sqrt{2}/\sqrt{5})\sin(\sqrt{6}Dt/\hbar) + \hat{T}^3_{\pm1}(i)[\tfrac{2}{5} + \tfrac{3}{5}\cos(\sqrt{6}Dt/\hbar)] \tag{7}$$

for $S = 3/2$ spin ensembles.

Secondly, we drop the terms oscillating at the high frequencies of $\sqrt{6}D/\hbar$ and $2\sqrt{6}D/\hbar$ in equation (4). Thus

$$\mathcal{H}'_{\rm int} = -\sqrt{5}g\mu_{\rm B}B_{\rm APP}\left\{\sum_i \hat{T}^1_0(i)\right\} - \tfrac{5}{2}\sum_{\{i,j\}}(J_{ij} + K_{ij})\hat{T}^1_0(i)\hat{T}^1_0(j)$$

$$+ \tfrac{5}{2}\sum_{\{i,j\}} J_{ij}\{[\hat{T}^1_1(i)\hat{T}^1_{-1}(j) + \hat{T}^1_{-1}(i)\hat{T}^1_1(j)](17/50)$$

$$+ [\hat{T}^2_1(i)\hat{T}^2_{-1}(j) + \hat{T}^2_{-1}(i)\hat{T}^2_1(j)](3/10)$$

$$+ [\hat{T}^3_1(i)\hat{T}^3_{-1}(j) + \hat{T}^3_{-1}(i)\hat{T}^3_1(j)](9/25)$$

$$- [\hat{T}^3_1(i)\hat{T}^1_{-1}(j) + \hat{T}^1_{-1}\hat{T}^3_1(j) + \hat{T}^1_1(i)\hat{T}^3_{-1}(j) + \hat{T}^3_{-1}(i)\hat{T}^1_1(j)](\sqrt{6}/50)\}. \tag{8}$$

Note the appearance of the rank 2 and rank 3 tensors in the exchange terms of equation (8). These are generated by the 'beating' of the Heisenberg exchange term with the single-ion quadrupole interaction.

Thirdly, now that the important exchange terms have been identified, we transform back to the laboratory frame. As with the $S = 1$ problem, we find that the truncated Hamiltonian of equation (8) is 'invariant' with respect to the quadrupolar transformation of equation (3). Thus in the laboratory frame the transformed Hamiltonian may be re-expressed in the form

$$\mathcal{H}_{\rm mod} = \mathcal{H}'_{\rm int} + \mathcal{H}_{\rm D} \tag{9}$$

where the full crystal field Hamiltonian $\mathcal{H}_{\rm D}$ has now been recovered.

Given the Hamiltonian of equation (9), Green's function theory can be employed to generate three coupled equations of motion:

$$
\begin{aligned}
E\langle\langle \hat{T}^1_1(l); \hat{T}^n_{-1}(m)\rangle\rangle = {} & a_1[n]\langle \hat{T}^n_0(m)\rangle \delta_{lm}/(2\pi) + g\mu_B B_{APP}\langle\langle \hat{T}^1_1(l); \hat{T}^n_{-1}(m)\rangle\rangle \\
& + \sqrt{5} \sum_{j\neq l} \{(J_{lj} + K_{lj})\langle\langle \hat{T}^1_1(l)\hat{T}^1_0(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& - 17/50 J_{lj}\langle\langle \hat{T}^1_0(l)\hat{T}^1_1(j); \hat{T}^n_{-1}(m)\rangle\rangle\} \\
& - 5\sum_{j\neq l} J_{lj}\{(3\sqrt{2}/10\sqrt{5})\langle\langle \hat{T}^2_2(l)\hat{T}^2_{-1}(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& + (3\sqrt{2}/10\sqrt{5})\langle\langle \hat{T}^2_0(l)\hat{T}^2_1(j); \hat{T}^n_{-1}(m)\rangle\rangle + (9/25)\langle\langle \hat{T}^3_2(l)\hat{T}^3_{-1}(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& + (9\sqrt{6}/25\sqrt{5})\langle\langle \hat{T}^3_0(l)\hat{T}^3_1(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& - (\sqrt{6}/50)\langle\langle \hat{T}^3_2(l)\hat{T}^1_{-1}(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& - (6/50\sqrt{5})\langle\langle \hat{T}^3_0(l)\hat{T}^1_1(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& - (\sqrt{6}/50\sqrt{5})\langle\langle \hat{T}^1_0(l)\hat{T}^3_1(j); \hat{T}^n_{-1}(m)\rangle\rangle\} \\
& + (3\sqrt{2}/\sqrt{5})D\langle\langle \hat{T}^2_1(l); \hat{T}^n_{-1}(m)\rangle\rangle
\end{aligned} \tag{10}
$$

$$
\begin{aligned}
E\langle\langle \hat{T}^2_1(l); \hat{T}^n_{-1}(m)\rangle\rangle = {} & \{a_2[n]\langle \hat{T}^{n-1}_0(m)\rangle + a_3[n]\langle \hat{T}^{n-1}_0(m)\rangle\}\delta_{lm}/(2\pi) \\
& + g\mu_B B_{APP}\langle\langle \hat{T}^2_1(l); \hat{T}^n_{-1}(m)\rangle\rangle \\
& + \sqrt{5} \sum_{j\neq l} \{(J_{lj} + K_{lj})\langle\langle \hat{T}^2_1(l)\hat{T}^1_0(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& + (2\sqrt{2}/5)J_{lj}\langle\langle \hat{T}^2_2(l)\hat{T}^1_{-1}(j); \hat{T}^n_{-1}(m)\rangle\rangle\} \\
& - 5\sum_{j\neq l} J_{lj}\{(15\sqrt{3}/50\sqrt{5})\langle\langle \hat{T}^2_0(l)\hat{T}^1_1(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& + (3/10\sqrt{5})\langle\langle \hat{T}^1_0(l)\hat{T}^2_1(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& + (10\sqrt{3}/25\sqrt{5})\langle\langle \hat{T}^2_2(l)\hat{T}^3_{-1}(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& + (15\sqrt{2}/50\sqrt{5})\langle\langle \hat{T}^2_0(l)\hat{T}^3_1(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& - (3/5\sqrt{5})\langle\langle \hat{T}^3_0(l)\hat{T}^2_1(j); \hat{T}^n_{-1}(m)\rangle\rangle\} \\
& + (3\sqrt{2}/\sqrt{5})D\langle\langle \hat{T}^1_1(l); \hat{T}^n_{-1}(m)\rangle\rangle + (2\sqrt{3}/\sqrt{5})D\langle\langle \hat{T}^3_1(l); \hat{T}^n_{-1}(m)\rangle\rangle
\end{aligned} \tag{11}
$$

$$
\begin{aligned}
E\langle\langle \hat{T}^3_1(l); \hat{T}^n_{-1}(m)\rangle\rangle = {} & \{a_4[n]\langle \hat{T}^{n-2}_0(m)\rangle + a_5[n]\langle \hat{T}^n_0(m)\rangle + a_6[n]\langle \hat{T}^{n+2}_0(m)\rangle\}\delta_{lm}/(2\pi) \\
& + g\mu_B B_{APP}\langle\langle \hat{T}^3_1(l); \hat{T}^n_{-1}(m)\rangle\rangle + (2\sqrt{3}/\sqrt{5})D\langle\langle \hat{T}^2_1(l); \hat{T}^n_{-1}(m)\rangle\rangle \\
& + \sqrt{5} \sum_{j\neq l} \{(J_{lj} + K_{lj})\langle\langle \hat{T}^3_1(l)\hat{T}^1_0(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& + (9\sqrt{6}/25)J_{lj}\langle\langle \hat{T}^3_0(l)\hat{T}^1_1(j); \hat{T}^n_{-1}(m)\rangle\rangle\} \\
& + 5\sum_{j\neq l} J_{lj}\{(17/50)\langle\langle \hat{T}^3_2(l)\hat{T}^1_{-1}(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& + (3\sqrt{3}/10\sqrt{5})\langle\langle \hat{T}^2_2(l)\hat{T}^2_{-1}(j); \hat{T}^n_{-1}(m)\rangle\rangle \\
& - (3\sqrt{2}/10\sqrt{5})\langle\langle \hat{T}^2_0(l)\hat{T}^2_1(j); \hat{T}^n_{-1}(m)\rangle\rangle
\end{aligned}
$$

**Table 1.**

| $n$ | $a_1[n]$ | $a_2[n]$ | $a_3[n]$ | $a_4[n]$ | $a_5[n]$ | $a_6[n]$ |
|---|---|---|---|---|---|---|
| 1 | $\frac{-1}{\sqrt{5}}$ | 0 | $\frac{-\sqrt{3}}{\sqrt{5}}$ | 0 | $\frac{-\sqrt{6}}{\sqrt{5}}$ | 0 |
| 2 | $\frac{-\sqrt{3}}{\sqrt{5}}$ | $\frac{-1}{\sqrt{5}}$ | $\frac{-2}{\sqrt{5}}$ | 0 | $\frac{-\sqrt{2}}{\sqrt{5}}$ | 0 |
| 3 | $\frac{-\sqrt{6}}{\sqrt{5}}$ | $\frac{-\sqrt{2}}{\sqrt{5}}$ | 0 | $\frac{-1}{\sqrt{5}}$ | $\frac{1}{\sqrt{5}}$ | 0 |

$$
\begin{aligned}
&- (9/25\sqrt{5})\langle\langle \hat{T}_0^1(l)\hat{T}_1^3(j); \hat{T}_{-1}^n(m)\rangle\rangle \\
&+ (12/25\sqrt{5})\langle\langle \hat{T}_0^3(l)\hat{T}_1^3(j); \hat{T}_{-1}^n(m)\rangle\rangle - (\sqrt{6}/50)\langle\langle \hat{T}_2^3(l)\hat{T}_{-1}^3(j); \hat{T}_{-1}^n(m)\rangle\rangle \\
&+ (\sqrt{6}/50\sqrt{5})\langle\langle \hat{T}_0^1(l)\hat{T}_1^1(j); \hat{T}_{-1}^n(m)\rangle\rangle\}
\end{aligned} \tag{12}
$$

where the coefficients $a_1[n]$, $a_2[n]$, $a_3[n]$, $a_4[n]$, $a_5[n]$ and $a_6[n]$, for $n = 1$, 2 and 3, are simple numerical coefficients defined in table 1. On implementing the RPA, and invoking translational invariance, equations (10)–(12) are transformed to the following three equations.

$$
\begin{aligned}
E\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle &= a_1[n]\langle \hat{T}_0^n(m)\rangle\delta_{lm}/(2\pi) + g\mu_B B_{APP}\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle \\
&+ \sqrt{5}\sum_{j\neq l}\langle \hat{T}_0^1\rangle\{(J_{lj} + K_{lj})\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle - (17/50)J_{lj}\langle\langle \hat{T}_1^1(j); \hat{T}_{-1}^n(m)\rangle\rangle\} \\
&- 5\sum_{j\neq l} J_{lj}\{(3\sqrt{2}/10\sqrt{5})\langle \hat{T}_0^2\rangle\langle\langle \hat{T}_1^2(j); \hat{T}_{-1}^n(m)\rangle\rangle \\
&+ (9\sqrt{6}/25\sqrt{5})\langle \hat{T}_0^3\rangle\langle\langle \hat{T}_1^3(j); \hat{T}_{-1}^n(m)\rangle\rangle \\
&- (\sqrt{6}/50\sqrt{5})\langle \hat{T}_0^1\rangle\langle\langle \hat{T}_1^3(j); \hat{T}_{-1}^n(m)\rangle\rangle - (6/50\sqrt{5})\langle \hat{T}_0^3\rangle\langle\langle \hat{T}_1^1(j); \hat{T}_{-1}^n(m)\rangle\rangle\} \\
&+ (3\sqrt{2}/\sqrt{5})D\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle
\end{aligned} \tag{13}
$$

$$
\begin{aligned}
E\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle &= \{a_2[n]\langle \hat{T}_0^{n-1}(m)\rangle + a_3[n]\langle \hat{T}_0^{n-1}(m)\rangle\}\delta_{lm}/(2\pi) \\
&+ g\mu_B B_{APP}\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle + \sqrt{5}\sum_{j\neq l}(J_{lj} + K_{lj})\langle \hat{T}_0^1\rangle\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle \\
&- 5\sum_{j\neq l} J_{lj}\{(15\sqrt{3}/50\sqrt{5})\langle \hat{T}_0^2\rangle\langle\langle \hat{T}_1^1(j); \hat{T}_{-1}^n(m)\rangle\rangle \\
&+ (3/10\sqrt{5})\langle \hat{T}_0^1\rangle\langle\langle \hat{T}_1^2(j); \hat{T}_{-1}^n(m)\rangle\rangle - (3/5\sqrt{5})\langle \hat{T}_0^3\rangle\langle\langle \hat{T}_1^2(j); \hat{T}_{-1}^n(m)\rangle\rangle \\
&+ (15\sqrt{2}/50\sqrt{5})\langle \hat{T}_0^2\rangle\langle\langle \hat{T}_1^3(j); \hat{T}_{-1}^n(m)\rangle\rangle\} + (3\sqrt{2}/\sqrt{5})D\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle \\
&+ (2\sqrt{3}/\sqrt{5})D\langle\langle \hat{T}_1^3(l); \hat{T}_{-1}^n(m)\rangle\rangle
\end{aligned} \tag{14}
$$

$$
\begin{aligned}
E\langle\langle\hat{T}_1^3(l);\hat{T}_{-1}^n(m)\rangle\rangle &= \{a_4[n]\langle\hat{T}_0^{n-2}(m)\rangle + a_5[n]\langle\hat{T}_0^n(m)\rangle + a_6[n]\langle\hat{T}_0^{n+2}(m)\rangle\}\delta_{lm}/(2\pi) \\
&+ g\mu_B B_{APP}\langle\langle\hat{T}_1^3(l);\hat{T}_{-1}^n(m)\rangle\rangle + (2\sqrt{3}/\sqrt{5})D\langle\langle\hat{T}_1^2(l);\hat{T}_{-1}^n(m)\rangle\rangle \\
&+ \sqrt{5}\sum_{j\neq l}\{(J_{lj}+K_{lj})\langle\hat{T}_0^1\rangle\langle\langle\hat{T}_1^3(l);\hat{T}_{-1}^n(m)\rangle\rangle \\
&+ (9\sqrt{6}/25)J_{lj}\langle\hat{T}_0^3\rangle\langle\langle\hat{T}_1^1(j);\hat{T}_{-1}^n(m)\rangle\rangle\} \\
&+ 5\sum_{j\neq l}J_{lj}\{-(3\sqrt{2}/10\sqrt{5})\langle\hat{T}_0^2\rangle\langle\langle\hat{T}_1^2(j);\hat{T}_{-1}^n(m)\rangle\rangle \\
&- (9/25\sqrt{5})\langle\hat{T}_0^1\rangle\langle\langle\hat{T}_1^3(j);\hat{T}_{-1}^n(m)\rangle\rangle + (12/25\sqrt{5})\langle\hat{T}_0^3\rangle\langle\langle\hat{T}_1^3(j);\hat{T}_{-1}^n(m)\rangle\rangle \\
&+ (\sqrt{6}/50\sqrt{5})\langle\hat{T}_0^1\rangle\langle\langle\hat{T}_1^1(j);\hat{T}_{-1}^n(m)\rangle\rangle\}. \qquad (15)
\end{aligned}
$$

Finally, on taking the spatial Fourier transforms of equations (13)–(15) we find

$$
\begin{aligned}
E(k)G_1 &= a_1[n]\langle\hat{T}_0^n\rangle/(2\pi) + [g\mu_B B_{APP} + \sqrt{5}\langle\hat{T}_0^1\rangle\{J(0)+K(0)\} - (\sqrt{5}/50)(17\langle\hat{T}_0^1\rangle \\
&- 6\langle\hat{T}_0^3\rangle)J(k)]G_1 + [(3\sqrt{2}/\sqrt{5})D - (3\sqrt{3}/2\sqrt{5})\langle\hat{T}_0^2\rangle J(k)]G_2 \\
&+ (\sqrt{6}/10\sqrt{5})(\langle\hat{T}_0^1\rangle - 18\langle\hat{T}_0^3\rangle)J(k)G_3 \qquad (16)
\end{aligned}
$$

$$
\begin{aligned}
E(k)G_2 &= \{a_2[n]\langle\hat{T}_0^{n-1}\rangle + a_3[n]\langle\hat{T}_0^{n+1}\rangle\}/(2\pi) \\
&+ [(3\sqrt{2}/\sqrt{5})D - (3\sqrt{3}/2\sqrt{5})\langle\hat{T}_0^2\rangle J(k)]G_1 \\
&+ [g\mu_B B_{APP} + \sqrt{5}\langle\hat{T}_0^1\rangle\{J(0)+K(0)\} - (3\sqrt{5}/10)(\langle\hat{T}_0^1\rangle + 2\langle\hat{T}_0^3\rangle)J)k)]G_2 \\
&+ [(2\sqrt{3}/\sqrt{5})D - (3/\sqrt{10})\langle\hat{T}_0^2\rangle J(k)]G_3 \qquad (17)
\end{aligned}
$$

$$
\begin{aligned}
E(k)G_3 &= \{a_4[n]\langle\hat{T}_0^{n-2}\rangle + a_5[n]\langle\hat{T}_0^n\rangle + a_6[n]\langle\hat{T}_0^{n+2}\rangle\}/(2\pi) \\
&+ [(2\sqrt{3}/\sqrt{5})D - (3/\sqrt{10})\langle\hat{T}_0^2\rangle J(k)]G_2 \\
&+ [g\mu_B B_{APP} + \sqrt{5}\langle\hat{T}_0^1\rangle\{J(0)+K(0)\} - (3\sqrt{5}/10)(3\langle\hat{T}_0^1\rangle - 4\langle\hat{T}_0^3\rangle)J(k)]G_3 \\
&+ (\sqrt{6}/10\sqrt{5})(\langle\hat{T}_0^1\rangle - 18\langle\hat{T}_0^3\rangle)J(k)G_3 \qquad (18)
\end{aligned}
$$

where

$$G_1 = \sum_m \langle\langle\hat{T}_1^1(l);\hat{T}_{-1}^n(m)\rangle\rangle \exp[-\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_l - \boldsymbol{R}_m)] \qquad (19)$$

$$G_2 = \sum_m \langle\langle\hat{T}_1^2(l);\hat{T}_{-1}^n(m)\rangle\rangle \exp[-\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_l - \boldsymbol{R}_m)] \qquad (20)$$

$$G_3 = \sum_m \langle\langle\hat{T}_1^3(l);\hat{T}_{-1}^n(m)\rangle\rangle \exp[-\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_l - \boldsymbol{R}_m)] \qquad (21)$$

and

$$J(k) = \sum_j J_{lj} \exp[\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{\delta}_{lj}] \qquad (22)$$

respectively. Thus the three poles of the Greens functions $G_1$, $G_2$ and $G_3$ are given by

$$
\begin{aligned}
E_1(k) &= g\mu_B B_{APP} + \sqrt{5}\langle\hat{T}_0^1\rangle\{J(0)+K(0)\} \\
&- 3\{(1/2\sqrt{5})\langle\hat{T}_0^1\rangle + \tfrac{1}{2}\langle\hat{T}_0^2\rangle + (1/\sqrt{5})\langle\hat{T}_0^3\rangle\}J(k) + \sqrt{6}D \qquad (23a)
\end{aligned}
$$

$$E_2(k) = g\mu_B B_{APP} + \surd 5\langle \hat{T}_0^1\rangle\{J(0) + K(0)\} - (2/\surd 5)\{\langle \hat{T}_0^1\rangle - 3\langle \hat{T}_0^3\rangle\}J(k) \tag{23b}$$

$$E_3(k) = g\mu_B B_{APP} + \surd 5\langle \hat{T}_0^1\rangle\{J(0) + K(0)\} - 3\{(1/2\surd 5)\langle \hat{T}_0^1\rangle - \tfrac{1}{2}\langle \hat{T}_0^2\rangle + (1/\surd 5)\langle \hat{T}_0^3\rangle\}J(k) - \surd 6 D. \tag{23c}$$

Note that the excitation energies are characterized by single-ion crystal field excitations in the limit $D \to \infty$, and spin waves in the limit $D \to 0$. In particular, when $D = 0$ and $T \to 0$ K, the usual spin wave result

$$E_1(k) \to g\mu_B B_{APP} + \tfrac{3}{2}\{J(0) + K(0) - J(k)\} \tag{24}$$

is obtained, where we have made use of the identities

$$\langle \hat{T}_0^1\rangle_{T=0} = 3/2\surd 5 \qquad \langle \hat{T}_0^2\rangle_{T=0} = \tfrac{1}{2} \qquad \langle \hat{T}_0^3\rangle_{T=0} = 1/2\surd 5. \tag{25}$$

In practice, equations (16)–(18) must be solved simultaneously to yield explicit solutions for $G_1$, $G_2$ and $G_3$. Once these have been determined, expressions for $\langle\langle T_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle$, $\langle\langle \hat{T}_1^2(l); T_{-1}^n(m)\rangle\rangle$ and $\langle\langle \hat{T}_1^3(l); \hat{T}_{-1}^n(m)\rangle\rangle$, and hence the correlation functions, can be obtained by invoking the inverse spatial Fourier transforms

$$\langle\langle \hat{T}_1^1(l); \hat{T}_{-1}^n(m)\rangle\rangle = N^{-1}\Sigma_k G_1 \exp[+\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_l - \boldsymbol{R}_m)] \tag{26}$$

$$\langle\langle \hat{T}_1^2(l); \hat{T}_{-1}^n(m)\rangle\rangle = N^{-1}\Sigma_k G_2 \exp[+\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_l - \boldsymbol{R}_m)] \tag{27}$$

$$\langle\langle \hat{T}_1^3(l); \hat{T}_{-1}^n(m)\rangle\rangle = N^{-1}\Sigma_k G_2 \exp[+\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_l - \boldsymbol{R}_m)] \tag{28}$$

and subsequently applying the spectral theorem (Zubarev 1960). In this way we obtain the self-correlation functions

$$\langle \hat{T}_{-1}^n \hat{T}_1^1\rangle = a_1[n]\langle \hat{T}_0^n\rangle\varphi_{11} + \{a_2[n]\langle \hat{T}_0^{n-1}\rangle + a_3[n]\langle \hat{T}_0^{n+1}\rangle\}\varphi_{12} + \{a_4[n]\langle \hat{T}_0^{n-2}\rangle + a_5[n]\langle \hat{T}_0^n\rangle + a_6[n]\langle \hat{T}_0^{n+2}\rangle\}\varphi_{13} \tag{29}$$

and

$$\langle \hat{T}_{-1}^n \hat{T}_1^2\rangle = a_1[n]\langle \hat{T}_0^n\rangle\varphi_{12} + \{a_2[n]\langle \hat{T}_0^{n-1}\rangle + a_3[n]\langle \hat{T}_0^{n+1}\rangle]\varphi_{22} + \{a_4[n]\langle \hat{T}_0^{n-2}\rangle + a_5[n]\langle \hat{T}_0^n\rangle + a_6[n]\langle \hat{T}_0^{n+2}\rangle\}\varphi_{23} \tag{30}$$

and

$$\langle \hat{T}_{-1}^n \hat{T}_1^3\rangle = a_1[n]\langle \hat{T}_0^n\rangle\varphi_{13} + \{a_2[n]\langle \hat{T}_0^{n-1}\rangle + a_3[n]\langle \hat{T}_0^{n+1}\rangle]\varphi_{23} + \{a_4[n]\langle \hat{T}_0^{n-2}\rangle + a_5[n]\langle \hat{T}_0^n\rangle + a_6[n]\langle \hat{T}_0^{n+2}\rangle\}\varphi_{33} \tag{31}$$

which is our principal result. In these equations, the $\varphi_{ij}$ are thermal weighting functions which depend implicitly on the excitation branches $E_1(k)$, $E_2(k)$ and $E_3(k)$. Using equations (16)–(18) and (23), we find that

$$\varphi_{11} = (1/10)(3\varphi_a + 4\varphi_b + 3\varphi_c) \tag{32a}$$

$$\varphi_{12} = (\surd 3/2\surd 5)(\varphi_a - \varphi_c) \tag{32b}$$

$$\varphi_{13} = (\surd 6/10)(\varphi_a - 2\varphi_b + \varphi_c) \tag{32c}$$

$$\varphi_{22} = (1/2)(\varphi_a + \varphi_c) \tag{32d}$$

$$\varphi_{23} = (1/\surd 10)(\varphi_a - \varphi_c) = (\surd 2/\surd 3)\varphi_{12} \tag{32e}$$

$$\varphi_{33} = (1/5)(\varphi_a + 3\varphi_b + \varphi_c) \tag{32f}$$

where the three functions $\varphi_a$, $\varphi_b$, and $\varphi_c$ are simply the occupation numbers for the three excitation branches taken separately:

$$\varphi_a = \frac{1}{N}\sum_k \left[\frac{1}{(\exp[\beta E_1(k)] - 1)}\right] \quad (33a)$$

$$\varphi_b = \frac{1}{N}\sum_k \left[\frac{1}{(\exp[\beta E_2(k)] - 1)}\right] \quad (33b)$$

$$\varphi_c = \frac{1}{N}\sum_k \left[\frac{1}{(\exp[\beta E_3(k)] - 1)}\right]. \quad (33c)$$

It is worth noting that the $\varphi_{ij}$ obtained from the TH/RPA results are just simple linear combinations of the individual occupation numbers $\varphi_a$, $\varphi_b$ and $\varphi_c$ for the three excitation branches. In the ordinary RPA, such simple expressions for the $\varphi_{ij}$ do not emerge.

Next, we note that the three self-correlation functions of equations (29)–(31) can be used, independently, to generate the set of ensemble averages $\langle\hat{T}_0^1\rangle, \langle\hat{T}_0^2\rangle$ and $\langle\hat{T}_0^3\rangle$. However, as with the TH/RPA treatment of the $S = 1$ ferromagnet, it is found that all three self-correlation functions yield the same, unique, ensemble averages:

$$\langle\hat{T}_0^1\rangle = \frac{(3 + 4\varphi_a + 3\varphi_b + 3\varphi_c + 3\varphi_a\varphi_b + 4\varphi_a\varphi_c + 3\varphi_b\varphi_c)}{2\sqrt{5}[1 + 2\varphi_a + \varphi_b + \varphi_c + 3\varphi_a\varphi_b + 2\varphi_a\varphi_c + \varphi_b\varphi_c + 4\varphi_a\varphi_b\varphi_c]} \quad (34)$$

$$\langle\hat{T}_0^2\rangle = \frac{(1 + \varphi_b + \varphi_c - \varphi_a\varphi_b + \varphi_b\varphi_c)}{2[1 + 2\varphi_a + \varphi_b + \varphi_c + 3\varphi_a\varphi_b + 2\varphi_a\varphi_c + \varphi_b\varphi_c + 4\varphi_a\varphi_b\varphi_c]} \quad (35)$$

$$\langle\hat{T}_0^3\rangle = \frac{(1 - 2\varphi_a + \varphi_b + \varphi_c + \varphi_a\varphi_b - 2\varphi_a\varphi_c + \varphi_b\varphi_c)}{2\sqrt{5}[1 + 2\varphi_a + \varphi_b + \varphi_c + 3\varphi_a\varphi_b + 2\varphi_a\varphi_c + \varphi_b\varphi_c + 4\varphi_a\varphi_b\varphi_c]} \quad (36)$$

irrespective of the size of $D$. In practice, of course, $\langle\hat{T}_0^1\rangle$, $\langle\hat{T}_0^2\rangle$ and $\langle\hat{T}_0^3\rangle$ must be calculated self-consistently through the use of equations (23) and (33)–(36).

## 4. Spin wave behaviour and energy gap considerations

In this section, we set limits on the regions where the TH/RPA model might be expected to hold. In the first place, if we set the crystal field parameter $D \equiv 0$, the three excitation branches reduce to

$$E_1(k) = g\mu_B B_{APP} + \sqrt{5}\langle\hat{T}_0^1\rangle\{J(0) + K(0)\} - 3\{(1/2\sqrt{5})\langle\hat{T}_0^1\rangle + \tfrac{1}{2}\langle\hat{T}_0^2\rangle + (1/\sqrt{5})\langle\hat{T}_0^3\rangle\}J(k) \quad (37a)$$

$$E_2(k) = g\mu_B B_{APP} + \sqrt{5}\langle\hat{T}_0^1\rangle\{J(0) + K(0)\} - (2/\sqrt{5})\{\langle\hat{T}_0^1\rangle - 3\langle\hat{T}_0^3\rangle\}J(k) \quad (37b)$$

$$E_3(k) = g\mu_B B_{APP} + \sqrt{5}\langle\hat{T}_0^1\rangle\{J(0) + K(0)\} - 3\{(1/2\sqrt{5})\langle\hat{T}_0^1\rangle - \tfrac{1}{2}\langle\hat{T}_0^2\rangle + (1/\sqrt{5})\langle\hat{T}_0^3\rangle\}J(k). \quad (37c)$$

In the limit $T = 0$ K, $\langle\hat{T}_0^1\rangle = (3/\surd 5)\langle\hat{T}_0^2\rangle = 3\langle\hat{T}_0^3\rangle$ and so the three excitation branches reduce to a single dispersive branch

$$E_1(k)_{T=0\,\mathrm{K}}^{D=0} = g\mu_B B_{\mathrm{APP}} + \tfrac{3}{2}\{J(0) + K(0) - J(k)\} \tag{38a}$$

and two non-dispersive branches

$$E_2(k)_{T=0\,\mathrm{K}}^{D=0} = g\mu_B B_{\mathrm{APP}} + \tfrac{3}{2}\{J(0) + K(0)\} \tag{38b}$$

$$E_3(k)_{T=0\,\mathrm{K}}^{D=0} = g\mu_B B_{\mathrm{APP}} + \tfrac{3}{2}\{J(0) + K(0)\}. \tag{38c}$$

The first of the three energy branches can be identified with the usual spin wave result. At absolute zero, only the $|I_z = 3/2\rangle$ ground state is populated, and so collective modes result from excitations involving ground states $|I_z = 3/2\rangle$ to first excited states $|I_z = 1/2\rangle$. The non-dispersive branches $E_2(k)$ and $E_3(k)$, on the other hand, correspond to single-ion excitations from $|I_z = 1/2\rangle$ to $|I_z = -1/2\rangle$ and $|I_z = -1/2\rangle$ to $|I_z = -3/2\rangle$, respectively, which are not populated at $T = 0$ K. However, as the temperature is raised, $\langle\hat{T}_0^2\rangle$ and $\langle\hat{T}_0^3\rangle$ will differ from their saturated values, and so three dispersive branches will emerge.

From an examination of equation (23), it is evident that the energy gap $\Delta(E_1(0))$, can be written in the form

$$\Delta = \surd 6D + g\mu_B B_{\mathrm{APP}} + \surd 5\langle\hat{T}_0^1\rangle K(0) + (1/2\surd 5)\{7\langle\hat{T}_0^1\rangle - 3\surd 5\langle\hat{T}_0^2\rangle - 6\langle\hat{T}_0^3\rangle\}J(0). \tag{39}$$

As noted earlier, the last term in equation (39) vanishes at $T = 0$ K, because $\langle\hat{T}_0^1\rangle = (3/\surd 5)\langle\hat{T}_0^2\rangle = 3\langle\hat{T}_0^3\rangle$ at saturation. However at higher temperatures, it is possible in the small $D$ limit that $\langle\hat{T}_0^2\rangle$ and $\langle\hat{T}_0^3\rangle$ will fall more rapidly than $\langle\hat{T}_0^1\rangle$. This will give rise to unphysical behaviour, in that the energy gap will increase with increasing temperature. Note however that, in the presence of a large crystal field parameter $D$ both $\langle\hat{T}_0^1\rangle$ and $\langle\hat{T}_0^3\rangle$ will decrease more quickly than $\langle\hat{T}_0^2\rangle$ with increasing temperature. Indeed at the Curie temperature, $\langle\hat{T}_0^2\rangle$ is necessarily finite, while $\langle\hat{T}_0^1\rangle$ and $\langle\hat{T}_0^3\rangle$ are identically zero. Thus for large $D$ the energy gap $\Delta$ will clearly 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 we can conclude that the TH/RPA model yields physically reasonable results when either $D/J(0) \geqslant 1$ or $K(0)/J(0) \geqslant 3$. This question is taken up again in the next section, where various estimates of the Curie temperature are compared and discussed.

## 5. The Curie temperature

In the situation where $D = 0$, Tahir-Khelli and ter Haar (1962) first showed that the Curie temperature $T_C$ for the $S = 3/2$ Heisenberg ferromagnet is given by

$$k_B T_C = \frac{5}{4}\left(\frac{1}{N}\sum_k \frac{1}{J(0) + K(0) - J(k)}\right)^{-1} \tag{40}$$

in the ordinary RPA. The molecular field (MF) prediction for $T_C$ is given by

$$k_B T_C = \frac{5}{4}\left(\frac{1}{N}\sum_k \frac{1}{J(0) + K(0)}\right)^{-1}. \tag{41}$$

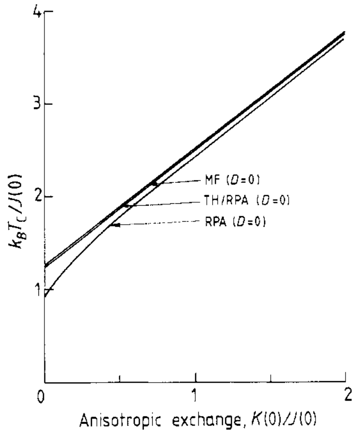


**Figure 1.** The Curie temperature as a function of the anisotropic exchange $K(0)$, for $D = 0$, as given by: the RPA (equation (40)), the MF (equation (41)) and the TH/RPA (equation (42)). These results have been obtained using a FCC cubic lattice, which of course cannot support an axial crystal field. Nevertheless we have chosen to use this lattice for comparison purposes.

For the TH/RPA however, we obtain the unique solution

$$k_BT_C = \frac{1}{4}\left[3\left(\frac{1}{N}\sum_k \frac{1}{J(0)+K(0)-0.3J(k)}\right)^{-1} + 2\left(\frac{1}{N}\sum_k \frac{1}{J(0)+K(0)-0.4J(k)}\right)^{-1}\right]. \tag{42}$$

A comparison of the calculated Curie temperatures for an FCC lattice, obtained using the RPA, MF and TH/RPA (equations (40), (41) and (42)) for various ratios of $K(0)/J(0)$ can be seen in figure 1. In the limit $K(0) \rightarrow \infty$, all the three estimates for $T_C$ coverge.

The situation for $D > 0$ is more complex because $\langle \hat{T}_0^2 \rangle$ is now finite at $T_C$. for the TH/RPA model we find, using equations (34) and (35), that

$$k_BT_C = \frac{6\langle \hat{T}_0^2\rangle}{(1-\theta_a+\theta_c)}\left[\frac{1}{N}\sum_k \frac{[J(0)+K(0)-0.3J(k)]}{2\{1-\cosh[\beta(\sqrt{6}D-1.5\langle \hat{T}_0^2\rangle J(k))]\}}\right] \tag{43}$$

where (i)

$$\langle \hat{T}_0^2\rangle = \frac{(1-\theta_a+\theta_c)}{2(1+3\theta_a+\theta_c+4\theta_a\theta_c)} \tag{44}$$

and (ii)

$$\theta_a = \frac{1}{N}\sum_k \{\exp[\beta(\sqrt{6}D-1.5\langle \hat{T}_0^2\rangle J(k))]-1\}^{-1} \tag{45a}$$

$$\theta_c = \frac{1}{N}\sum_k \{\exp[-\beta(\sqrt{6}D-1.5\langle \hat{T}_0^2\rangle J(k))\Big]-1\}^{-1}. \tag{45b}$$

As noted earlier the TH/RPA result gives physically reasonable results at finite temperatures, when $D/J(0) \geqslant 1$.

Since $\langle \hat{T}_0^2\rangle$ is temperature dependent it is necessary to solve for both $\langle \hat{T}_0^2\rangle$ and $k_BT_C$ simultaneously in equations (43)–(45). The computed values of $T_C$ (for $K(0) = 0$) in the TH/RPA model can be seen in figure 2, as a function of $D/J(0)$. From an examination of this diagram it will be observed that $D \rightarrow \infty$ the predicted value of $T_C$, obtained using the TH/RPA, saturates at $0.982J(0)$ for a FCC lattice. This is not entirely unexpected since molecular field theory predicts a limit of $T_C = J(0)$. As $D \rightarrow \infty$, the crystal field ground

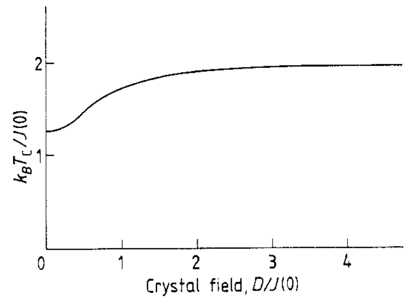


**Figure 2.** The Curie temperature as a function of the crystal field parameter $D$, for $K(0) = 0$, as given by the TH/RPA (equation (43)).

**Table 2.** Predicted Curie temperatures $k_BT_C/J(0)$.

| (*a*) $D \to \infty$ ($K(0) = 0$) | | | | |
|---|---|---|---|---|
| Lattice | Series (Ising doublet) [Fisher (1967)] | TH/RPA | Mean field theory | RPA |
| FCC | 0.8162 | 0.982 | 1.0 | 0.0 |
| (*b*) $D \to 0$ ($K(0) = 0$) | | | | |
| Lattice | Series (Heisenberg $S = 1$) [Fisher (1967)] | TH/RPA | Mean field theory | RPA |
| FCC | 0.767 | 0.985 | 1.0 | 0.765 |

*Note*: (i) The results have been scaled so that the mean field results are unity. (ii) The Tahir-Kheli and ter Haar values agree remarkably well with the high-temperature series results for $D = 0$. (iii) The ordinary RPA predicts $T_C \to 0$ as $D \to \infty$, which is unphysical.

state reduces to a $|S_z = \pm 3/2\rangle$ doublet. Consequently, the magnetic exchange terms are only effective within the $|\pm 3/2\rangle$ doublet, with little influence from the high-energy $|\pm 1/2\rangle$ states.

Finally, in table 2 the predicted Curie temperatures for the TH/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)) and the high-temperature series results (see for example the review by Fisher (1967)). It will be observed that the Curie temperatures of the TH/RPA, for small $D$, are disappointingly close to the mean field results. In practice, however, 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.

## 6. Equivalent two-parameter single-particle density operator

Callen and Shtrikman (1965) first showed that in the absence of crystal fields, the higher-order moments calculated from a many-body treatment of the Heisenberg ferromagnet can be mimicked by a temperature-renormalized single-parameter single-ion density matrix

$$\rho = \frac{\exp(-xS_z)}{\mathrm{Tr}[\exp(-xS_z)]}. \tag{46}$$

This density matrix yields exactly the same ensemble averages as the many-body treatment, provided $x$ is chosen such that

$$\langle S_z\rangle_{\mathrm{mb}} = \langle S_z\rangle_{\mathrm{sp}}[=\sqrt{5}\langle\hat{T}_0^1\rangle_{S=3/2}]. \tag{47}$$

Recently however, Bowden and Martin (1990b) have shown that for the $S = 1$ easy axis ferromagnet, it is possible to construct a two-parameter single-particle density matrix

$$\rho = \frac{\exp\{xS_z + (y/\sqrt{6})[3S_z^2 - S(S+1)]\}}{\mathrm{Tr}[\exp\{xS_z + (y/\sqrt{6})[3S_z^2 - S(S+1)]\}]} \tag{48}$$

which reproduces the same ensemble averages $\langle\hat{T}_0^1\rangle$ and $\langle\hat{T}_0^2\rangle$ as those calculated using the TH/RPA. The need for a second parameter arises because the axially symmetric crystal field gives rise to two excitation branches $E_1(k)$ and $E_2(k)$. For spin 3/2 ensembles, there are three excitation branches $E_1(k)$, $E_2(k)$ and $E_3(k)$. Thus it is of some interest to enquire whether or not a two-parameter single-particle density matrix can still be constructed which mimics the TH/RPA results.

For $S = 3/2$, the single-ion model of equation (48) is characterized by three $\Delta m = \pm 1$ transitions

$$E_{|3/2\rangle} = E_{|1/2\rangle} = (x + \sqrt{6}y)k_{\mathrm{B}}T \tag{49a}$$

$$E_{|1\cdot 2\rangle} - E_{|-1/2\rangle} = xk_{\mathrm{B}}T \tag{49b}$$

$$E_{|-1/2\rangle} - E_{|-3/2\rangle} = (x - \sqrt{6}y)k_{\mathrm{B}}T \tag{49c}$$

which are the single ion counterparts of the three excitation branches $E_1(k)$, $E_2(k)$ and $E_3(k)$. In order to make contact with the TH/RPA, we define $x$ and $y$ via the occupation numbers of the corresponding excitation branch

$$\frac{1}{\exp\{x + \sqrt{6}y\} - 1} = \varphi_a = \frac{1}{N}\sum_k \frac{1}{\exp(\beta E_1(k)) - 1} \tag{50a}$$

$$\frac{1}{\exp\{x\} - 1} = \varphi_b = \frac{1}{N}\sum_k \frac{1}{\exp(\beta E_2(k)) - 1} \tag{50b}$$

$$\frac{1}{\exp\{x - \sqrt{6}y\} - 1} = \varphi_c = \frac{1}{N}\sum_k \frac{1}{\exp(\beta E_3(k)) - 1}. \tag{50c}$$

This ensures that the single-ion description possesses the same thermal occupation numbers as that of the TH/RPA. Next, on substituting equations (57)–(59) into the many-body expressions for $\langle\hat{T}_0^1\rangle$, $\langle\hat{T}_0^2\rangle$ and $\langle\hat{T}_0^3\rangle$ (equations (36)–(38)) it can be shown, after some manipulation, that

$$\langle\hat{T}_0^1\rangle = \frac{3\exp(3x/2 + \sqrt{6}y/2) + \exp(x/2 - \sqrt{6}y/2) - \exp(-x/2 - \sqrt{6}y/2) - 3\exp(-3x/2 + \sqrt{6}y/2)}{2\sqrt{5}[\exp(3x/2 + \sqrt{6}y/2) + \exp(x/2 - \sqrt{6}y/2) + \exp(-1x/2 - \sqrt{6}y/2) + \exp[-3x/2 + 6y/2]} \tag{51}$$

$$\langle\hat{T}_0^2\rangle = \frac{\exp(3x/2 + \sqrt{6}y/2) - \exp(x/2 - \sqrt{6}y/2) - \exp(-x/2 - \sqrt{6}y/2) + \exp(-3x/2 + \sqrt{6}y/2)}{2[\exp(3x/2 + \sqrt{6}y/2) + \exp(x/2 - \sqrt{6}y/2) + \exp(-x2 - \sqrt{6}y/2) + \exp(-3x/2 + \sqrt{6}y/2)]} \tag{52}$$

$$\langle \hat{T}_0^3 \rangle = \frac{\begin{array}{l}\exp(3x/2 + \sqrt{6}y/2) - 3\exp(x/2 - \sqrt{6}y/2) \\ \qquad + 3\exp(-x/2 - \sqrt{6}y/2) - \exp(-3x/2 + \sqrt{6}y/2)\end{array}}{\begin{array}{l}2\sqrt{5}[\exp(3x/2 + \sqrt{6}y/2) + \exp(x/2 - \sqrt{6}y/2) \\ \qquad + \exp(-x/2 - \sqrt{6}y/2) + \exp(-3x/2 + \sqrt{6}y/2)]\end{array}}. \tag{53}$$

These results are identical to the single-ion averages obtained using equation (48). Thus the temperature-renormalized effective single-particle density matrix operator of equation (48), yields the same ensemble averages as the many-body TH/RPA for both $S = 1$ and $3/2$ spin ensembles.

## 7. Conclusions

In this paper it has been shown that large axially symmetric quadrupole crystal field interactions can be incorporated into a many-body treatment of the easy axis Heisenberg $S = 3/2$ ferromagnet, using the transformed Hamiltonian/random phase approximation (TH/RPA). In addition, explicit expressions have been given for (i) the ensemble averages $\langle \hat{T}_0^n \rangle$, and (ii) the Curie temperature $T_C$. Within the context of the TH/RPA these expressions are 'unique' in marked contrast to earlier work. Finally, a single-particle model, in the spirit of Callen and Shtrikman, has been constructed which yields exactly the same ensemble averages as the TH/RPA for $S = 3/2$ easy axis ferromagnetic ensembles.

## References

Bowden G J and Martin J P D 1990a *J. Phys.: Condens. Matter* **2** 10435–50
—— 1990b *J. Phys.: Condens. Matter* **2** 10451–60
Bowden G J, Martin J P D and Oitmaa J 1986 *J. Phys. C: Solid State Phys.* **19** 551–61
Buckmaster H A, Chatterjee R and Singh Y H 1972 *Phys. Status Solidi* **13** 9–50
Callen H B 1963 *Phys. Rev.* **130** 890
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
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 *Rep. Prog. Phys.* 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
Tahir-Kheli R 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