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Temperature dependence of the single-ion lattice anisotropy coefficients of a ferromagnetic Heisenberg monolayer

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Abstract. In investigations of the orientation of the magnetization of thin ferromagnetic films, the single-ion anisotropy coefficients play a crucial role. By applying a thermodynamic perturbation theory we calculate the temperature dependence of the second- and fourth-order single-ion anisotropies for a Heisenberg monolayer with Tyablikov decoupling (the random-phase approximation, RPA) and compare with results obtained from mean-field theory. In order to assess the accuracy of the Tyablikov (RPA) and also the Callen decoupling approximations in the Green's function many-body theory, we calculate the magnetization of a Heisenberg spin pair and of a ferromagnetic monolayer, and compare the results with exact solutions available for the spin pair with arbitrary spins, and a recent quantum Monte Carlo calculation (for spin 1/2) for the monolayer. The RPA decoupling provides a fairly good approximation to the exact results for the magnetization over the whole temperature range of interest. Because of this, we expect the calculated anisotropy coefficients to be approximated well by this method.

1. Introduction

New experimental results concerning the temperature dependence of the magnetic anisotropy and the resulting orientation of the magnetization of thin films and multilayer systems [1] have provided an incentive for theoretical investigations [2]. Because of its feasibility, the mean-field theory (MFT) for a Heisenberg Hamiltonian plus anisotropy terms has been primarily applied; for a review see [3]. In such two-dimensional (2D) systems, however, correlations beyond MFT should be taken into account, e.g. by using many-body Green's function theory [4, 5], which is capable of treating the problem over the whole temperature range of interest. In this study, we apply this theory in order to calculate the temperature dependence of the second- and fourth-order single-ion anisotropy coefficients of a ferromagnetic Heisenberg monolayer using the Tyablikov decoupling (the random-phase approximation, RPA) [6].

We start by examining the accuracy of the RPA, and also going beyond the RPA, for the Callen decoupling [7] for two different spin systems for which exact solutions for the magnetization are available. First we consider the Heisenberg spin pair. For this case we calculate the magnetization for arbitrary spins S_1 and S_2. Secondly, we treat the Heisenberg monolayer with $S = 1/2$ spins. For this system, a recent quantum Monte Carlo (QMC) calculation [8] can be used as an 'exact' reference for the corresponding results obtained from the RPA and Callen decoupling. In both systems, the Heisenberg exchange coupling and an external magnetic field are taken into account. Results for the magnetization calculated using the MFT are also presented for comparison.

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As the main subject of the paper we present results of calculations of the temperature dependence of the second- and fourth-order single-ion anisotropy coefficients for the Heisenberg monolayer (with $S = 2$ and $S = 10$ spins as examples), and compare the results obtained with RPA and MFT. Because the result for the magnetization calculated with RPA is, in contrast to that from MFT, very close to the exact result, we expect that the anisotropy coefficients calculated with RPA to also be quite accurate and to represent a considerable improvement over the MFT results.

2. The Heisenberg spin pair

The Hamiltonian for the Heisenberg spin pair with spins S_1 and S_2 with exchange coupling constant J in an external magnetic field $B = Be_z$ reads

$$H = -B(S_1^z + S_2^z) - \frac{J}{2}(S_1 \cdot S_2 + S_2 \cdot S_1). \tag{1}$$

In order to calculate the magnetization

$$\langle S^z \rangle = -\partial F / \partial B \tag{2}$$

from the free energy $F = -\beta^{-1} \ln Z$, with the inverse temperature $\beta = 1/(k_B T)$ and k_B the Boltzmann constant, one has to determine the partition function

$$Z = \text{Tr} \exp(-\beta H) = \sum_i \exp(-\beta \, \varepsilon_i)$$

from the eigenvalues ε_i of the Hamiltonian matrix

$$\langle S_1, S_2, S_T, M_T | H | S_1, S_2, S_T, M_T \rangle. \tag{3}$$

Using the relation $2S_1 \cdot S_2 = S_T^2 - S_1^2 - S_2^2$, the $(2S_1 + 1)(2S_2 + 1)$ eigenvalues $\varepsilon(S_T, M_T)$ are determined by

$$\varepsilon(S_T, M_T) = -\frac{J}{2}\Big(S_T(S_T + 1) - S_1(S_1 + 1) - S_2(S_2 + 1)\Big) - BM_T \tag{4}$$

where the total spin quantum number is denoted by S_T with values running from $|S_1 - S_2|$ to $S_1 + S_2$, and the total magnetic quantum number is $M_T = M_1 + M_2 = -S_T, \ldots, S_T$.

The exact expression for the total magnetization is then obtained from equation (2):

$$\langle S^z \rangle = \left(\sum_{S_T} \sum_{M_T} M_T e^{-\beta \varepsilon(S_T, M_T)} \right) \Big/ \left(\sum_{S_T} \sum_{M_T} e^{-\beta \varepsilon(S_T, M_T)} \right). \tag{5}$$

As an example we consider the case where $S_1 = S_2 = 1$. The $(2S_1 + 1)(2S_2 + 1) = 9$ eigenvalues are obtained from equation (4), and the exact analytical result for the magnetization per spin reads

$$\langle S^z \rangle = \frac{2 \sinh(2\beta B) \exp(\beta J) + 2 \cosh(\beta J) \sinh(\beta B)}{2 \exp(\beta J) \cosh(2\beta B) + 4 \cosh(\beta J) \cosh(\beta B) + \exp(-2\beta J) + \exp(\beta J)}. \tag{6}$$

Next we compare the exact magnetization of the two-spin system with that obtained from many-body Green's function techniques (RPA and Callen decoupling). We use the set of Green's functions in the energy representation [5]

$$G_{ij}^{(n)} = \langle\langle S_i^+; (S_j^z)^n S_j^- \rangle\rangle \qquad i, j = 1, 2 \tag{7}$$

with the respective equations of motion

$$\omega \langle\langle S_i^+; (S_j^z)^n S_j^- \rangle\rangle = A_{ij}^{(n)} + \langle\langle [S_i^+, H]; (S_j^z)^n S_j^- \rangle\rangle. \tag{8}$$

The inhomogeneities are given by

$$A_{ij}^{(n)} = A_i^{(n)} \delta_{ij} = \langle [S_i^+, (S_j^z)^n S_j^-] \rangle$$
$$A_i^{(n)} = 2\langle (S_i^z - 1)^n S_i^z \rangle + \langle \{(S_i^z - 1)^n - (S_i^z)^n\}\{S(S+1) - S_i^z - (S_i^z)^2\} \rangle. \tag{9}$$

Evaluating the commutator $[S_i^+, H]$ occurring on the right-hand side of equation (8) leads to higher-order Green's functions which have to be approximated. In the case of a homogeneous system, i.e. where $\langle S_i^z \rangle = \langle S^z \rangle$, the RPA consists in the decoupling [6]

$$\langle\langle (S_i^z S_j^+ - S_i^+ S_j^z); (S_i^z)^n S_i^- \rangle\rangle \approx \langle S^z \rangle (G_{ji}^{(n)} - G_{ii}^{(n)}). \tag{10}$$

The equations of motion then reduce to a set of linear eigenvalue equations from which the Green's functions can be determined. For simplicity we focus on the case where $S_1 = S_2$. Then we have to consider only the Green's functions $G_{11}^{(n)}$ and $G_{21}^{(n)}$, and the equations determining the Green's functions read

$$\begin{pmatrix} \omega - B - J\langle S^z \rangle & J\langle S^z \rangle \\ J\langle S^z \rangle & \omega - B - J\langle S^z \rangle \end{pmatrix} \begin{pmatrix} G_{11}^{(n)} \\ G_{21}^{(n)} \end{pmatrix} = \begin{pmatrix} A_1^{(n)} \\ 0 \end{pmatrix}. \tag{11}$$

To determine the magnetization $\langle S^z \rangle$ we need $G_{11}^{(n)}$. From equation (11) one derives

$$G_{11}^{(n)} = \frac{1}{2} \sum_{i=1,2} \frac{A_1^{(n)}}{\omega - \omega_i} \qquad \text{with } \omega_1 = B \text{ and } \omega_2 = B + 2J\langle S^z \rangle. \tag{12}$$

For spin S one obtains a set of coupled equations for $n = 0, 1, \ldots, 2S - 1$ from the spectral theorem [4]:

$$\langle (S_1^z)^n S_1^- S_1^+ \rangle = S(S+1)\langle (S_1^z)^n \rangle - \langle (S_1^z)^{n+1} \rangle - \langle (S_1^z)^{n+2} \rangle$$
$$= \lim_{\delta \to 0} \frac{i}{2\pi} \int d\omega \frac{1}{e^{\beta\omega} - 1} (G_{11}^{(n)}(\omega + i\delta) - G_{11}^{(n)}(\omega - i\delta))$$
$$= \frac{1}{2} A_1^{(n)} \sum_i (e^{\beta\omega_i} - 1)^{-1} \tag{13}$$

where the largest moments $\langle (S_1^z)^{2S+1} \rangle$ can be expressed in terms of lower moments with the help of the relation

$$\prod_{r=-S}^{r=+S} (S_1^z - r) = 0.$$

For the case of spin $S = 1$, one has to solve the equations for $n = 0$ and $n = 1$ from which the RPA result for the magnetization $\langle S^z \rangle$ is obtained:

$$\langle S^z \rangle_{S=1}^{RPA} = \frac{1 + m_1 + m_2}{1 + \frac{3}{2}(m_1 + m_2) + \frac{3}{4}(m_1 + m_2)^2} \tag{14}$$

with

$$m_1 = (e^{\beta B} - 1)^{-1} \qquad \text{and} \qquad m_2 = (e^{\beta(B+2J\langle S^z \rangle)} - 1)^{-1}. \tag{15}$$

Now we perform an analogous calculation with the decoupling procedure proposed by Callen [7], which replaces equation (10) by

$$\langle\langle (S_i^z S_j^z - S_i^+ S_j^z); (S_i^z)^n S_i^- \rangle\rangle \approx \langle S^z \rangle (G_{ji}^{(n)} - G_{ii}^{(n)}) - \alpha \big(\langle (S_i^z)^n S_i^- S_j^+ \rangle G_{ii}^{(n)} - \langle S_j^- (S_i^z)^n S_i^z \rangle G_{ji}^{(n)} \big) \tag{16}$$

with $\alpha = \langle S^z \rangle / 2S^2$. For $\alpha = 0$, one recovers the RPA decoupling.

Using this decoupling in the equations of motion and determining the Green's functions and the correlation functions, we obtain a system of coupled equations for expectation values which for spin $S = 1$ reads

$$\langle S^z \rangle = \left(3\langle (S^z)^2 \rangle - \langle S^z \rangle - 2\right) \frac{b_1 m_1^1 + a_1 m_2^1}{a_1 + b_1} + \langle (S^z)^2 \rangle \tag{17}$$

$$\langle (S^z)^2 \rangle = 2 - \langle S^z \rangle (1 + m_1^0 + m_2^0) \tag{18}$$

$$\langle S_1^- S_2^+ \rangle = \langle S^z \rangle (m_1^0 - m_2^0) \tag{19}$$

$$\langle S_1^z S_1^- S_2^+ \rangle = \left(3\langle (S^z)^2 \rangle - \langle S^z \rangle - 2\right) \frac{a_1 (m_1^1 - m_2^1)}{a_1 + b_1} \tag{20}$$

where

$$m_1^0 = m_1^1 = \left(e^{\beta B} - 1\right)^{-1} \tag{21}$$

$$m_2^0 = \left(e^{\beta(B + J\langle S^z \rangle (2 + \langle S_1^- S_2^+ \rangle))} - 1\right)^{-1} \tag{22}$$

$$m_2^1 = \left(e^{\beta(B + J\langle S^z \rangle (2 + \langle S_1^z S_1^- S_1^+ \rangle + \langle S_1^- S_2^+ \rangle / 2))} - 1\right)^{-1} \tag{23}$$

$$a_1 = J\langle S^z \rangle \left(1 + \frac{1}{2}\langle S_1^z S_1^- S_2^+ \rangle\right) \tag{24}$$

$$b_1 = J\langle S^z \rangle \left(1 + \frac{1}{2}(\langle S_1^z S_1^- S_2^+ \rangle + \langle S_1^- S_2^+ \rangle)\right). \tag{25}$$

These equations have been solved numerically.

In figure 1 we compare the exact results for the magnetization as a function of the temperature for the $S_1 = S_2 = 1$ case, equation (6), with those for the RPA, the Callen
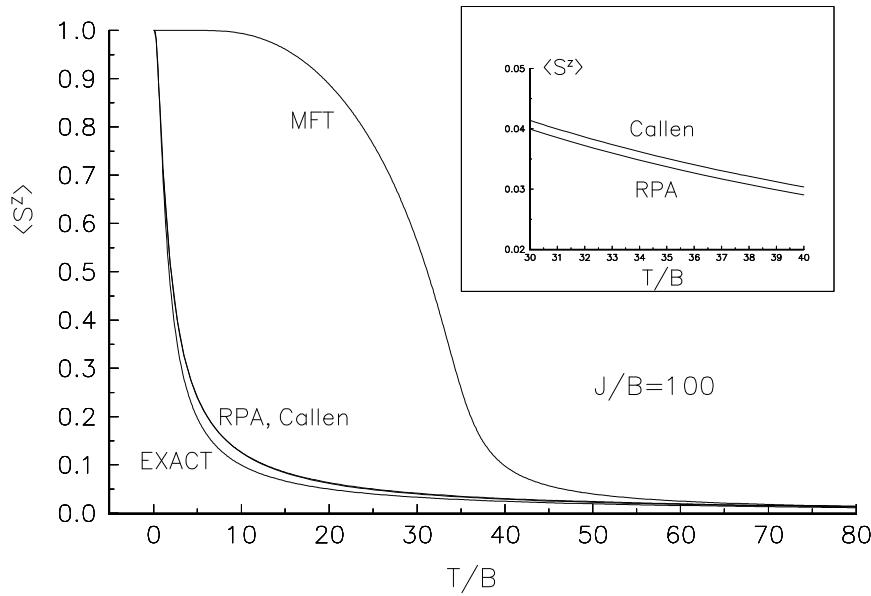

Figure 1. The temperature dependence of the magnetization $\langle S^z \rangle$ of the Heisenberg spin pair with $S_1 = S_2 = 1$. The exact result, equation (6), is compared with that from mean-field theory (MFT), the RPA, and the Callen decoupling. The RPA and the Callen results are indistinguishable within the line thickness of the figure. By amplifying the scale we show, in the inset, that the RPA is slightly better than the Callen decoupling. We have used $J/B = 100$.

decoupling, and for the mean-field theory. The latter reads

$$\langle S^z \rangle_{S=1}^{MFT} = \frac{1+m}{1+3\,m+3\,m^2} \qquad \text{with } m = \left(e^{\beta(B+J\langle S^z \rangle/2)} - 1 \right)^{-1}. \tag{26}$$

We use $J = 100$ for the exchange coupling constant, and $B = 1$ for the magnetic field. These values correspond to a rare-earth ferromagnet in an external magnetic field of about 1 T.

One observes that the RPA and Callen approximations are fairly close to the exact result and yield a substantial improvement over MFT. Despite the fact that additional correlations are taken into account in the Callen decoupling, the result of the latter is very close to and practically indistinguishable within the line thickness from the RPA result. With an amplified scale, the inset in figure 1 shows that the result of the Callen decoupling is unexpectedly slightly worse than that of the RPA. This is, however, consistent with the finding of Callen [7] that his procedure is superior to RPA only for higher spins when calculating the Curie temperature of a bulk ferromagnet.

One may apply the methods described above to calculate the magnetization of larger Heisenberg spin clusters of a given size and shape. An exact calculation of the magnetization is prohibitively difficult in this case. However, the Green's function theory is still a tractable method since it leads to a linear set of eigenvalue equations which can be solved numerically. The method allows one to take into account approximately the spin correlations over the whole temperature range of interest. We emphasize that the magnetization of such cluster systems including the collective excitations has been calculated up to now by use of the Holstein–Primakoff approximation [9], which is valid for low temperatures only.

3. The Heisenberg monolayer of a square lattice for $S = 1/2$

In this section we calculate the magnetization of a Heisenberg monolayer with spin $S = 1/2$ for a square lattice with RPA, Callen decoupling, and MFT. We use as an 'exact' reference a recent quantum Monte Carlo (QMC) calculation [8]. Before the latter result was published it was not possible to check the quality of the various approximations for the present case.

The Hamiltonian of the Heisenberg monolayer with a magnetic field reads

$$H_0 = -B \sum_l S_l^z - \frac{J}{2} \sum_{\langle kl \rangle} \boldsymbol{S}_k \cdot \boldsymbol{S}_l \tag{27}$$

where $\langle kl \rangle$ means summation over nearest neighbours only. An applied magnetic field and/or magnetic anisotropies will induce a long-range magnetic order with a Curie temperature of the order of magnitude of the exchange coupling [10]. Note that, owing to the transverse magnetic fluctuations with long wavelengths, an *isotropic* 2D Heisenberg magnet does not exhibit a finite magnetization at finite temperatures (the Mermin–Wagner theorem [11]).

In order to calculate the magnetization $\langle S_i^z \rangle$ for the case of spin $S = 1/2$:

$$\langle S_i^z \rangle = 1/2 - \langle S_i^- S_i^+ \rangle \tag{28}$$

the correlation function $\langle S_i^- S_i^+ \rangle$ is obtained from the Green's function $G_{ij} = \langle\langle S_i^+; S_j^- \rangle\rangle$ by using the spectral theorem; see equation (12). Within the RPA and the Callen decoupling, the Green's function in momentum space is given by

$$G_k(\omega) = \frac{2\langle S^z \rangle}{\omega - E_k^\alpha} \tag{29}$$

with the magnon dispersion relation

$$E_k^\alpha = B + \langle S^z \rangle J (4 - 2\gamma_k) \left(1 + \frac{\alpha}{N} \sum_k \gamma_k \coth(\beta E_k^\alpha / 2) \right). \tag{30}$$

For a square lattice (lattice constant $a_0 = 1$), $\gamma_k = \cos k_x + \cos k_y$, and N is the number of lattice sites. The x- and y-axes determine the monolayer plane. The RPA decoupling corresponds to $\alpha = 0$, and the Callen decoupling to $\alpha = 2\langle S^z \rangle$ [7]; see equation (16).

Inserting equations (29) and (30) into equation (28) yields an equation for the magnetization $\langle S^z \rangle$, which has to be solved self-consistently:

$$\langle S^z \rangle = 1/2 - \langle S^z \rangle \frac{1}{\pi^2} \int_0^\pi \mathrm{d}k_x \int_0^\pi \mathrm{d}k_y \, (\coth(\beta E_k^\alpha / 2) - 1) \tag{31}$$

where the sum over the momenta, $\sum_k$, has been replaced by an integral over the first Brillouin zone.

In figure 2 we compare the 'exact' quantum Monte Carlo calculation [8] of the magnetization for the spin-$S = 1/2$ monolayer with results obtained from RPA and Callen decoupling. For comparison we also show the MFT results. Following reference [8], we use the parameters $J = 4, 10, 20$, and $B = 1$. We find that the RPA calculations are fairly close to the QMC results. In the present case, the RPA is definitely closer to the exact result than the result obtained from the Callen decoupling, which is expected to improve for higher spins [7]. For higher spins, however, QMC calculations are not available as a reference. Both RPA and Callen calculations give a substantial improvement over MFT. We also state that the Green's function RPA result yields a better overall agreement with the QMC results than the magnetization calculated with the Schwinger boson approximations [8], which are, however, obtained for a continuous monolayer and not for a discrete lattice.

4. Temperature dependence of the lattice anisotropy coefficients for a monolayer in MFT and the RPA

Because the RPA outlined in the previous sections is already close to the exact results for the magnetization, we calculate the temperature-dependent (effective) lattice anisotropy coefficients of a square layer using RPA. These coefficients are accessible experimentally by measuring the orientation of the magnetization of thin films, for example. They are defined as the coefficients in an expansion of the free energy in powers of the cosine of θ [3], which is the polar angle between the magnetization $\langle S \rangle$ and the normal to the film surface:

$$F(T, \theta) = F_0(T) - \mathcal{K}_2(T) \cos^2 \theta - \mathcal{K}_4(T) \cos^4 \theta - \boldsymbol{B} \cdot \langle \boldsymbol{S} \rangle. \tag{32}$$

In reference [3], the effective anisotropy coefficients have been calculated within the mean-field theory for the magnetization. The total Hamiltonian $H = H_0 + V$ is separated into an unperturbed part H_0 consisting of the exchange coupling and the magnetic field (cf. equation (27)), and a perturbation V which denotes either the second- or the fourth-order single-ion lattice anisotropy:

$$V = -K_n \sum_l (S_l^z)^n \qquad n = 2, 4. \tag{33}$$

Within first-order thermodynamic perturbation theory (expansion in $1/k_B T$) the $\mathcal{K}_n(T)$ are given by

$$\mathcal{K}_n(T) = K_n f_n(T) \tag{34}$$

where the temperature dependence is introduced solely by the functions $f_n(T)$ which are expressed in terms of expectation values $\langle (S^z)^n \rangle_0$ with respect to the unperturbed Hamiltonian H_0 [3, 12]:

$$f_2(T) = \big(3\langle (S^z)^2 \rangle_0 - S(S+1)\big)/2 \tag{35}$$

$$f_4(T) = \big[35\langle (S^z)^4 \rangle_0 - \big(30S(S+1) - 25\big)\langle (S^z)^2 \rangle_0 + 3S(S+1)\big(S(S+1) - 2\big)\big]/8. \tag{36}$$
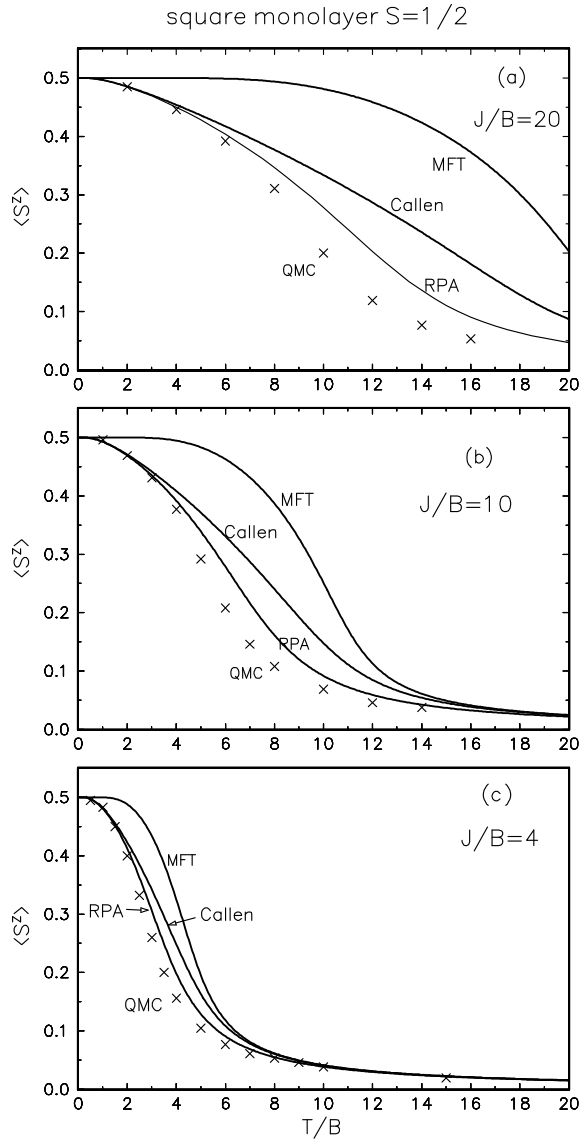
Figure 2. The temperature dependence of the magnetization $\langle S^z \rangle$ of a Heisenberg monolayer for a square lattice with $S = 1/2$. Comparison is made between the 'exact' quantum Monte Carlo (QMC) result [8] and the results obtained with MFT, the RPA, and Callen decoupling. We have used (a) $J/B = 20$, (b) $J/B = 10$, and (c) $J/B = 4$.

To demonstrate the significant change in the temperature dependence of the anisotropy coefficients, we compare the results obtained from the mean-field theory (MFT, reference [3]) and from the RPA decoupling for a Heisenberg monolayer of a square lattice. In the latter case, the magnetization $\langle S^z \rangle$ [13] and the moments $\langle (S^z)^n \rangle_0$ ($n = 2, 4$) are calculated from the RPA Green's function for spin S as described in [5]. One obtains

$$\langle S^z \rangle = \frac{[S - \varphi(S)][1 + \varphi(S)]^{2S+1} + [1 + S + \varphi(S)][\varphi(S)]^{2S+1}}{[1 + \varphi(S)]^{2S+1} - [\varphi(S)]^{2S+1}} \tag{37}$$

$$\langle (S^z)^2 \rangle_0 = S(S+1) - \langle S^z \rangle (1 + 2\,\varphi(S)) \tag{38}$$

$$\langle (S^z)^4 \rangle_0 = S(S+1)\big[8\,\varphi^2(S) + 8\,\varphi(S) + S(S+1)\big]$$
$$- \langle S^z \rangle \big[24\,\varphi^3(S) + 36\,\varphi^2(S) + \big(14 + 4S(S+1)\big)\varphi(S) + 2S(S+1) + 1\big] \tag{39}$$

where

$$\varphi(S) = \frac{1}{N} \sum_k \big(e^{\beta E_k^{\alpha=0}} - 1\big)^{-1} \tag{40}$$

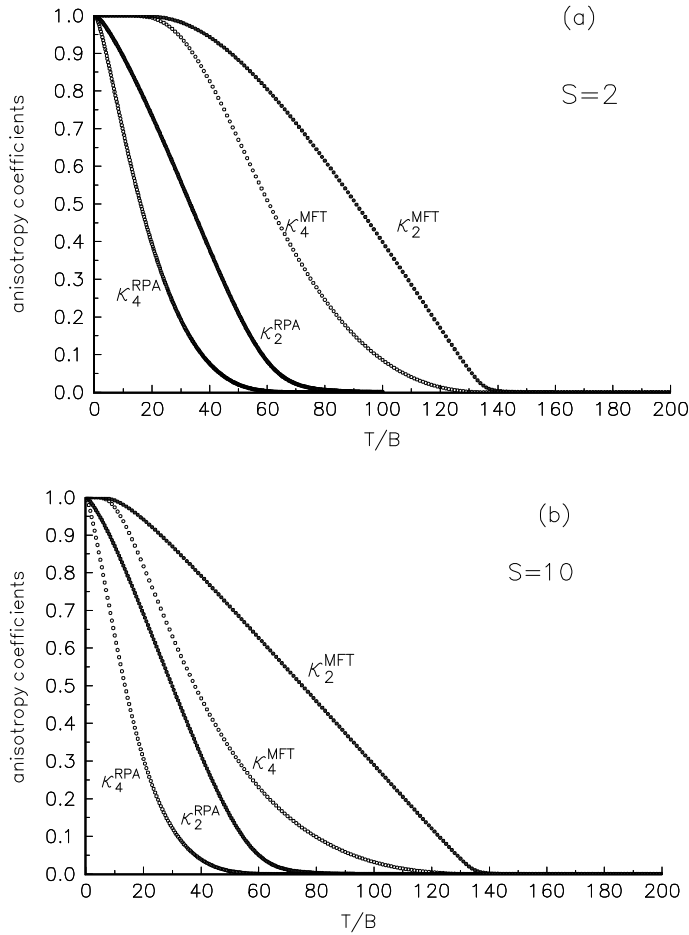where $E_k^{\alpha=0}$ is the RPA dispersion relation, equation (30).

Figure 3. The temperature dependence of the effective lattice anisotropy coefficients $\mathcal{K}_2(T)$ and $\mathcal{K}_4(T)$ of a square Heisenberg monolayer calculated with thermodynamic perturbation theory. The magnetization is determined with MFT and the RPA. We have used $J/B = 100$, and (a) $S = 2$, (b) $S = 10$. To allow for a comparison between different spin values, the interaction strengths are scaled according to $J \rightarrow J/S(S+1)$ and $B \rightarrow B/S$.

In figure 3 we compare the temperature-dependent anisotropy coefficients $\mathcal{K}_n(T)$, equation (34), for $S = 2$ and $S = 10$ calculated with the MFT and RPA. We use $K_n = 1$, and in order to obtain an appropriate scaling for different spins we put $J \rightarrow J/S(S+1)$ and $B \rightarrow B/S$. The resulting behaviour of the $\mathcal{K}_n(T)$ calculated by the RPA differs markedly from

that obtained from MFT, particularly at low temperatures. Whereas the $\mathcal{K}_n(T)$ as obtained within MFT show an exponential decay in this temperature range, those calculated using the RPA decrease more rapidly and exhibit a nearly linear behaviour. One also observes from figure 3 that the $\mathcal{K}_n(T)$ calculated with the RPA decoupling exhibit a much weaker dependence on the spin value S than those calculated with MFT.

5. Conclusions

In order to assess quantitatively the accuracy of Green's function decouplings, we have calculated the magnetization for a Heisenberg Hamiltonian consisting of an isotropic exchange interaction and an applied magnetic field for two systems for which exact solutions are available. In particular, a spin pair with arbitrary spin quantum numbers S (we have used $S_1 = S_2 = 1$ as an example) as well as a single square layer with $S = 1/2$ were investigated. We have demonstrated that the RPA and the Callen decoupling of a many-body Green's function theory represent an acceptable approximation to the exact results. Both approaches furnish a much better description of the magnetization than the mean-field approximation, which is expected to yield poor results for such low-dimensional systems. For the case of the Heisenberg monolayer, we have used as a reference the results obtained from a quantum Monte Carlo calculation [8]. In this case, the Callen decoupling yields worse results than the RPA decoupling, even though the former includes more correlations. This is consistent with the fact that the magnetic ordering temperature T_c for bulk lattices is better approximated by the RPA than by the Callen decoupling *for low spins S*. However, for larger spins S, the latter method yields improved values for T_c [7]. Thus, to test the accuracy of these different approaches for a thin film for general spins S it would be highly desirable to perform QMC calculations for larger S. Note, however, that although the Callen decoupling does not seem to yield better results for the magnetization as compared to the computationally simpler RPA method, it exhibits an improved description of the free energy (even for $S = 1/2$) at elevated temperatures [14], because it yields results closer to the paramagnetic limit.

Because of its importance for the orientation of the magnetization, we have calculated the temperature dependence of the anisotropy coefficients, $\mathcal{K}_n(T)$, by use of a thermodynamic perturbation theory. The fairly good approximation of the magnetization calculated with the RPA decoupling as shown by the comparison with the exact results of the QMC method (cf. section 3) suggests that the calculated effective anisotropy coefficients are also well approximated by applying the RPA. It turned out that the temperature dependence of the anisotropy coefficients calculated by using the RPA shows significant differences from that calculated using the MFT, in particular at low temperatures. A second important result is that the spin dependence of the $\mathcal{K}_n(T)$ is much weaker in the RPA than in MFT.

Note that in using thermodynamic perturbation theory the consideration of an applied magnetic field is essential, since an isotropic 2D Heisenberg magnet does not produce ordering at finite temperatures, in contrast to a 3D system [11]. The magnetization, and thus the effective anisotropies, depend on the magnetic field. We emphasize that without an applied field, the magnetic anisotropy cannot be handled as a small perturbation. In this case, the anisotropies, which consist of the lattice anisotropy and the long-range magnetic dipole coupling, must be treated on an equal footing with the exchange coupling. In order to determine the $\mathcal{K}_n(T)$, the free energy, $F(T, \theta)$ in equation (32), has to be calculated as a function of temperature and polar angle θ. For this purpose, the Heisenberg spins have to be rotated by the angle θ, and appropriate decoupling procedures for the Green's functions occurring have to be applied, e.g. along the lines proposed in reference [15]. Work in this direction is in progress.

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