

# Many-body Green's function theory for thin ferromagnetic anisotropic Heisenberg films: treatment of the exchange anisotropy

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**Abstract.** The many-body Green's function theory developed in our previous work for treating the reorientation of the magnetization of thin ferromagnetic films is extended to include the exchange anisotropy. This leads to additional momentum dependencies which require some non-trivial changes in the formalism. The theory is developed for arbitrary spin values  $S$  and for multilayers. The effects of the exchange anisotropy and the single-ion anisotropy, which was treated in our earlier work, on the magnetic properties of thin ferromagnetic films are compared.

**PACS.** 75.10.Jm Quantized spin models – 75.30.Ds Spin waves – 75.70.Ak Magnetic properties of monolayers and thin films

## 1 Introduction

There is increasing activity in experimental and theoretical investigations of thin magnetic films and multilayers. Of particular interest is the reorientation of the magnetization as function of temperature and film thickness. The simplest approach for treating thin ferromagnetic films is the application of mean field theory (MFT) to a Heisenberg model, by either diagonalizing the corresponding single-particle Hamiltonian [1] or applying thermodynamic perturbation theory [2]. This approximation completely neglects collective excitations (magnons = spin waves).

In order to take the influence of these collective excitations into account, we have turned to many-body Green's function theory (GFT), which allows reliable calculations over the entire range of temperature of interest. In reference [3] we treated a spin  $S = 1/2$  Heisenberg monolayer in a magnetic field and found, by comparing with Quantum Monte Carlo (QMC) calculations, that a Tyablikov (RPA) [4] decoupling is a very good approximation. Therefore, we did not try to go beyond RPA in the subsequent paper [5], in which we treated the field-induced magnetic reorientation of a ferromagnetic  $S = 1$  monolayer, whereby a second-order single-ion anisotropy was also included. Whereas the exchange interaction terms are decoupled by RPA, this is not allowed for the terms coming from the single-ion anisotropy because this leads to

unphysical results. Instead, we applied a decoupling procedure proposed by Anderson and Callen [6], which, however, is a good approximation only for small anisotropies. This was shown to be the case in reference [7], where we were able to treat the single-ion anisotropy exactly (for any strength) by introducing higher-order Green's functions and subsequently taking advantage of relations between products of spin operators, which leads to an automatic closure of the hierarchy of the equations of motion for the Green's functions with respect to the anisotropy terms; the terms from the exchange interaction are still decoupled by a generalized RPA scheme. In reference [8] we have investigated the quality of this approach by comparing with QMC calculations. In reference [7] we treated the spin  $S = 1$  case only; the formal generalization to spins  $S > 1$  is possible, but its numerical realisation is quite cumbersome. This is not the case when remaining at the level of the lowest-order Green's functions and applying the Anderson-Callen decoupling. In this case, not only is the treatment of spins  $S > 1$  feasible, but also multilayers can be described, as was done in reference [9]. To make the treatment of multilayers practicable, we had to apply a new method which not only uses the eigenvalues but also the eigenvectors of the non-symmetric matrix which governs the equations of motion for the Green's functions. We mention a few related papers in which Green's function theory is also applied to spin reorientation problems. In reference [10] Green's functions are applied to the reorientation problem after a Holstein-Primakoff mapping to bosons, which is only a valid description at low temperatures. In reference [11] the reorientation is obtained by

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the competition of an approximately decoupled single-ion anisotropy and a schematic shape anisotropy. In [12] the interplay of the dipole coupling with an easy-plane single-ion anisotropy is discussed, and in [13] the in-plane dipole coupling anisotropy of a square ferromagnetic Heisenberg monolayer is considered.

In our previous work mentioned above, we treated an isotropic Heisenberg exchange interaction plus a single-ion anisotropy, the magnetic dipole coupling, and an external magnetic field. In the present paper, we include the exchange anisotropy in the formalism and discuss its effect on the magnetic properties of thin ferromagnetic films, in particular in comparison to the influence of the single-ion anisotropy. As will be shown, the treatment of the exchange anisotropy requires some non-trivial changes in the formalism, which are necessary due to additional momentum dependencies which are absent in the treatment of the single-ion anisotropies.

The paper is organized as follows. In Section 2 we establish the Green's function formalism. For pedagogical reasons, a large part of the formalism is demonstrated for the monolayer case and subsequently generalised to the multilayer case, which can easily be done. In Section 3, we discuss the field-induced reorientation of the magnetization when using the exchange anisotropy for determining the orientation at temperature  $T = 0$ . We fix the strength of the exchange anisotropy in such a way that it produces the same Curie temperature for a monolayer as the application of the single-ion anisotropy strength used in our previous work [5,9]. This enables a comparison between the roles of the single-ion and exchange anisotropies on the magnetic properties of thin ferromagnetic films. Section 4 contains a summary of the results.

## 2 The Green's function formalism

We formulate the theory in such a way that the results of our previous work [5,9] are obtained as limiting cases.

We consider a spin Hamiltonian consisting of an isotropic Heisenberg exchange interaction with strength  $J_{kl}$  between nearest neighbour lattice sites, an exchange anisotropy with strength  $D_{kl}$ , a second-order single-ion lattice anisotropy with strength  $K_{2,k}$ , a magnetic dipole coupling with strength  $g_{kl}$ , and an external magnetic field  $\mathbf{B} = (B^x, B^y, B^z)$ :

$$\begin{aligned} \mathcal{H} = & -\frac{1}{2} \sum_{\langle kl \rangle} J_{kl} (S_k^- S_l^+ + S_k^z S_l^z) \\ & - \frac{1}{2} \sum_{\langle kl \rangle} D_{kl} S_k^z S_l^z - \sum_k K_{2,k} (S_k^z)^2 \\ & - \sum_k \left( \frac{1}{2} B^- S_k^+ + \frac{1}{2} B^+ S_k^- + B^z S_k^z \right) \\ & + \frac{1}{2} \sum_{kl} \frac{g_{kl}}{r_{kl}^5} \left( r_{kl}^2 (S_k^- S_l^+ + S_k^z S_l^z) - 3(\mathbf{S}_k \mathbf{r}_{kl})(\mathbf{S}_l \mathbf{r}_{kl}) \right). \end{aligned} \quad (1)$$

Here the notation  $S_k^\pm = S_k^x \pm iS_k^y$  and  $B^\pm = B^x \pm iB^y$  is introduced, where  $k$  and  $l$  are lattice site indices and  $\langle kl \rangle$  indicates summation over nearest neighbours only. The only difference from reference [9] is the additional exchange-anisotropy term.

In order to treat the reorientation problem for general spin  $S$ , we need the following Green's functions

$$G_{ij,\eta}^{\alpha,mn}(\omega) = \langle \langle S_i^\alpha; (S_j^z)^m (S_j^-)^n \rangle \rangle_{\omega,\eta}, \quad (2)$$

where  $\alpha = (+, -, z)$  takes care of all directions in space,  $\eta = \pm 1$  refers to the anticommutator or commutator Green's functions, respectively, and  $n \geq 1, m \geq 0$  are positive integers, necessary for dealing with higher spin values  $S$ .

The exact equations of motion are

$$\omega G_{ij,\eta}^{\alpha,mn}(\omega) = A_{ij,\eta}^{\alpha,mn} + \langle \langle [S_i^\alpha, \mathcal{H}]_-; (S_j^z)^m (S_j^-)^n \rangle \rangle_{\omega,\eta} \quad (3)$$

with the inhomogeneities

$$A_{ij,\eta}^{\alpha,mn} = \langle [S_i^\alpha, (S_j^z)^m (S_j^-)^n]_\eta \rangle, \quad (4)$$

where  $\langle \dots \rangle = \text{Tr}(\dots e^{-\beta \mathcal{H}})$ . The equations, leaving out for the moment the terms due to the dipole coupling, are given explicitly by

$$\begin{aligned} \omega G_{ij,\eta}^{\pm,mn} &= A_{ij,\eta}^{\pm,mn} \\ &\mp \sum_k J_{ik} \left( \langle \langle S_i^z S_k^\pm; (S_j^z)^m (S_j^-)^n \rangle \rangle \right. \\ &\quad \left. - \langle \langle S_k^z S_i^\pm; (S_j^z)^m (S_j^-)^n \rangle \rangle \right) \\ &\pm \sum_k D_{ik} \langle \langle S_k^z S_i^\pm; (S_j^z)^m (S_j^-)^n \rangle \rangle \\ &\pm K_{2,i} \langle \langle (S_i^\pm S_i^z + S_i^z S_i^\pm); (S_j^z)^m (S_j^-)^n \rangle \rangle \\ &\mp B^\pm G_{ij,\eta}^{z,mn} \pm B^z G_{ij,\eta}^{\pm,mn} \\ \omega G_{ij,\eta}^{z,mn} &= A_{ij(\eta)}^{z,mn} \\ &+ \frac{1}{2} \sum_k J_{ik} \langle \langle (S_i^- S_k^+ - S_k^- S_i^+); (S_j^z)^m (S_j^-)^n \rangle \rangle \\ &- \frac{1}{2} B^- G_{ij,\eta}^{+,mn} + \frac{1}{2} B^+ G_{ij,\eta}^{-,mn}. \end{aligned} \quad (5)$$

After solving these equations the components of the magnetization can be determined from the Green's functions *via* the spectral theorem. A solution is possible by establishing a closed system of equations by decoupling the higher-order Green's functions on the right hand sides. Contrary to reference [7], where we proceed to higher-order Green's functions, we stay here at the level of the lowest-order equations. For the exchange-interaction and exchange-anisotropy terms, we use a generalized Tyablikov- (or RPA-) decoupling

$$\begin{aligned} \langle \langle S_i^\alpha S_k^\beta; (S_j^z)^m (S_j^-)^n \rangle \rangle_\eta &\simeq \\ &\langle S_i^\alpha \rangle G_{kj,\eta}^{\beta,mn} + \langle S_k^\beta \rangle G_{ij,\eta}^{\alpha,mn}. \end{aligned} \quad (6)$$

The terms from the single-ion anisotropy have to be decoupled differently, because an RPA decoupling leads to unphysical results; *e.g.* for spin  $S = 1/2$ , the terms due to the single-ion anisotropy do not vanish in RPA, as they should do, because in this case  $\sum_i K_{2,i} \langle (S_i^z)^2 \rangle$  is a constant and should not influence the equations of motion. In the appendix of reference [5] we investigated different decoupling schemes proposed in the literature, *e.g.* those of Lines [14] or that of Anderson and Callen [6], which should be reasonable for single-ion anisotropies small compared to the exchange interaction. We found the Anderson-Callen decoupling to be most adequate. It consists in implementing the suggestion of Callen [15] to improve the RPA by treating the diagonal terms arising from the single-ion anisotropy as well. This leads to

$$\langle \langle (S_i^\pm S_i^z + S_i^z S_i^\pm); (S_j^z)^m (S_j^-)^n \rangle \rangle_\eta \simeq 2 \langle S_i^z \rangle \left( 1 - \frac{1}{2S^2} [S(S+1) - \langle S_i^z S_i^z \rangle] \right) G_{ij,\eta}^{\pm,mn}. \quad (7)$$

This term vanishes for  $S = 1/2$  as it should.

After a Fourier transform to momentum space, one obtains, for a ferromagnetic film with  $N$  layers,  $3N$  equations of motion for a  $3N$ -dimensional Green's function vector  $\mathbf{G}^{mn}$ :

$$(\omega \mathbf{1} - \mathbf{\Gamma}) \mathbf{G}^{mn} = \mathbf{A}^{mn}, \quad (8)$$

where  $\mathbf{1}$  is the  $3N \times 3N$  unit matrix. The Green's function vectors and inhomogeneity vectors each consist of  $N$  three-dimensional subvectors which are characterized by the layer indices  $i$  and  $j$

$$\mathbf{G}_{ij}^{mn}(\mathbf{k}, \omega) = \begin{pmatrix} G_{ij}^{+,mn}(\mathbf{k}, \omega) \\ G_{ij}^{-,mn}(\mathbf{k}, \omega) \\ G_{ij}^{z,mn}(\mathbf{k}, \omega) \end{pmatrix}, \quad \mathbf{A}_{ij}^{mn} = \begin{pmatrix} A_{ij}^{+,mn} \\ A_{ij}^{-,mn} \\ A_{ij}^{z,mn} \end{pmatrix}. \quad (9)$$

The equations of motion are then expressed in terms of these layer vectors, and  $3 \times 3$  submatrices  $\mathbf{\Gamma}_{ij}$  of the  $3N \times 3N$  matrix  $\mathbf{\Gamma}$

$$\left[ \omega \mathbf{1} - \begin{pmatrix} \mathbf{\Gamma}_{11} & \mathbf{\Gamma}_{12} & \dots & \mathbf{\Gamma}_{1N} \\ \mathbf{\Gamma}_{21} & \mathbf{\Gamma}_{22} & \dots & \mathbf{\Gamma}_{2N} \\ \dots & \dots & \dots & \dots \\ \mathbf{\Gamma}_{N1} & \mathbf{\Gamma}_{N2} & \dots & \mathbf{\Gamma}_{NN} \end{pmatrix} \right] \begin{bmatrix} \mathbf{G}_{1j} \\ \mathbf{G}_{2j} \\ \dots \\ \mathbf{G}_{Nj} \end{bmatrix} = \begin{bmatrix} \mathbf{A}_{1j} \delta_{1j} \\ \mathbf{A}_{2j} \delta_{2j} \\ \dots \\ \mathbf{A}_{Nj} \delta_{Nj} \end{bmatrix}, \quad j = 1, \dots, N. \quad (10)$$

After applying the decoupling procedures (6) and (7), the  $\mathbf{\Gamma}$  matrix reduces to a band matrix with zeros in the  $\mathbf{\Gamma}_{ij}$  sub-matrices, when  $j > i + 1$  and  $j < i - 1$ . The diagonal sub-matrices  $\mathbf{\Gamma}_{ii}$  are of size  $3 \times 3$  and have the form

$$\mathbf{\Gamma}_{ii} = \begin{pmatrix} H_i^z & 0 & -H_i^+ \\ 0 & -H_i^z & H_i^- \\ -\frac{1}{2} \tilde{H}_i^- & \frac{1}{2} \tilde{H}_i^+ & 0 \end{pmatrix} \quad (11)$$

where

$$\begin{aligned} H_i^z &= Z_i + \langle S_i^z \rangle J_{ii} (q - \gamma_{\mathbf{k}}), \\ Z_i &= B_i^z + D_{ii} q \langle S_i^z \rangle + (J_{i,i+1} + D_{i,i+1}) \langle S_{i+1}^z \rangle \\ &\quad + (J_{i,i-1} + D_{i,i-1}) \langle S_{i-1}^z \rangle \\ &\quad + K_{2,i} 2 \langle S_i^z \rangle \left( 1 - \frac{1}{2S^2} [S(S+1) - \langle S_i^z S_i^z \rangle] \right), \\ \tilde{H}_i^\pm &= B_i^\pm + \langle S_i^\pm \rangle J_{ii} (q - \gamma_{\mathbf{k}}) + J_{i,i+1} \langle S_{i+1}^\pm \rangle \\ &\quad + J_{i,i-1} \langle S_{i-1}^\pm \rangle, \\ H_i^\pm &= \tilde{H}_i^\pm - \langle S_i^\pm \rangle D_{ii} \gamma_{\mathbf{k}}. \end{aligned} \quad (12)$$

For a square lattice and a lattice constant taken to be unity,  $\gamma_{\mathbf{k}} = 2(\cos k_x + \cos k_y)$ , and  $q = 4$  is the number of intra-layer nearest neighbours. Except for the exchange-anisotropy terms,  $D_{ij}$ , these equations are the same as in reference [9]. Putting all  $D_{ij} = 0$ , one has  $\tilde{H}_i^\pm = H_i^\pm$ . Note that owing to the momentum dependence in  $H_i^\pm$  coming from the exchange anisotropy,  $\tilde{H}_i^\pm \neq H_i^\pm$ , which forbids a naive extension of the formalism of reference [9], as discussed below.

Approximating the dipole coupling by mean field theory (MFT), which is a good approximation when the dipole coupling strength is small as compared to the exchange interaction strength (as proved in Appendix A of [9]), one finds that the effects of the dipole coupling can be included as an effective field:

$$\begin{aligned} B_i^\pm &= B^\pm + \sum_{j=1}^N g_{ij} \langle S_j^\pm \rangle T^{|i-j|}, \\ B_i^z &= B^z - 2 \sum_{j=1}^N g_{ij} \langle S_j^z \rangle T^{|i-j|}, \end{aligned} \quad (13)$$

where the lattice sums for a two-dimensional square lattice are given by ( $n = |n - j|$ )

$$T^n = \sum_{lm} \frac{l^2 - n^2}{(l^2 + m^2 + n^2)^{5/2}}. \quad (14)$$

The indices  $lm$  run over all sites of the  $j$ th layer excluding terms with  $l^2 + m^2 + n^2 = 0$ .

One observes that the dipole coupling in MFT leads to a renormalization of the external field: there is an enhancement of the transverse fields, and a reduction of the field perpendicular to the film.

The  $3 \times 3$  off-diagonal sub-matrices  $\mathbf{\Gamma}_{ij}$  for  $j = i \pm 1$  are of the form

$$\mathbf{\Gamma}_{ij} = \begin{pmatrix} -J_{ij} \langle S_i^z \rangle & 0 & (J_{ij} + D_{ij}) \langle S_i^+ \rangle \\ 0 & J_{ij} \langle S_i^z \rangle & -(J_{ij} + D_{ij}) \langle S_i^- \rangle \\ \frac{1}{2} J_{ij} \langle S_i^- \rangle - \frac{1}{2} J_{ij} \langle S_i^+ \rangle & & 0 \end{pmatrix}. \quad (15)$$

When treating the monolayer, one can use the spectral theorem for calculating the components of the magnetization. This was done in reference [5] for the case of spin  $S = 1$  and the single-ion anisotropy by using the

commutator Green's functions. In order to obtain sufficient equations it was necessary, to add equations coming from the condition that the commutator Green's functions have to be regular at  $\omega = 0$ , which we call the regularity conditions.

For the multilayer problem, however, a naive application of the spectral theorem turned out to be forbiddingly difficult. Instead we invented a method, which we call the eigenvector method, that uses the eigenvectors as well as the eigenvalues of the  $\Gamma$ -matrix governing the equations of motion. This opened up a practicable way to treat multilayers [9].

If instead, anticommutator Green's functions are used, it is not necessary to introduce the regularity conditions, which nevertheless are valid. We demonstrate this explicitly for the monolayer. The use of the anticommutator Green's functions also suggests a way for finding the procedure which can deal with the additional  $\mathbf{k}$ -dependencies coming from the exchange anisotropy.

For simplicity, we consider the reorientation in the  $x$ - $z$ -plane, *i.e.* we use as external field  $\mathbf{B} = (B^x, 0, B^z)$ . The equations of motion for the monolayer in this case are

$$\begin{pmatrix} \omega - H^z & 0 & H^x \\ 0 & \omega + H^z & -H^x \\ +\frac{1}{2}\tilde{H}^x & -\frac{1}{2}\tilde{H}^x & \omega \end{pmatrix} \begin{pmatrix} G_{\mathbf{k},\eta}^{+,mn} \\ G_{\mathbf{k},\eta}^{-,mn} \\ G_{\mathbf{k},\eta}^{z,mn} \end{pmatrix} = \begin{pmatrix} A_{\mathbf{k},\eta}^{+,mn} \\ A_{\mathbf{k},\eta}^{-,mn} \\ A_{\mathbf{k},\eta}^{z,mn} \end{pmatrix}. \quad (16)$$

This system of equations has three eigenvalues

$$\omega_1 = 0; \quad \omega_{2,3} = \pm\epsilon_{\mathbf{k}} = \pm\sqrt{H^z H^z + \tilde{H}^x H^x}. \quad (17)$$

and the equations are solved by

$$G_{\mathbf{k},\eta}^{\alpha,mn} = \frac{\Delta_{\eta}^{\alpha,mn}}{\Delta}, \quad (18)$$

where  $\Delta_{\eta}^{\alpha,mn}$  is the determinant of the matrix in equation (16) where column  $\alpha$  is replaced by the inhomogeneity vector, and  $\Delta = \omega(\omega - \epsilon_{\mathbf{k}})(\omega + \epsilon_{\mathbf{k}})$ .

Now the spectral theorem [16] is applied in momentum space

$$C_{\mathbf{k}}^{\alpha,mn} = \langle (S^z)^m (S^-)^n S^{\alpha} \rangle_{\mathbf{k}} = \lim_{\delta \rightarrow 0} \frac{i}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega}{e^{\beta\omega} + 1} \times \left( G_{\mathbf{k},\eta=+1}^{\alpha,mn}(\omega + i\delta) - G_{\mathbf{k},\eta=+1}^{\alpha,mn}(\omega - i\delta) \right). \quad (19)$$

Using the relation between anticommutator and commutator

$$A_{\mathbf{k},\eta=+1}^{\alpha,mn} = A_{\eta=-1}^{\alpha,mn} + 2C_{\mathbf{k}}^{\alpha,mn}, \quad (20)$$

where it is important that the commutator inhomogeneities  $A_{\eta=-1}^{\alpha,mn}$  do not depend on the momentum  $\mathbf{k}$ , one

obtains the following set of equations

$$H^z C_{\mathbf{k}}^{+,mn} - H^x C_{\mathbf{k}}^{z,mn} = A_{\eta=-1}^{+,mn} \left( \frac{1}{2}\epsilon_{\mathbf{k}} \coth\left(\frac{\beta\epsilon_{\mathbf{k}}}{2}\right) - \frac{1}{2}H^z \right) + \frac{1}{2}H^x A_{\eta=-1}^{z,mn}, \quad (21)$$

$$-H^z C_{\mathbf{k}}^{-,mn} + H^x C_{\mathbf{k}}^{z,mn} = A_{\eta=-1}^{-,mn} \left( \frac{1}{2}\epsilon_{\mathbf{k}} \coth\left(\frac{\beta\epsilon_{\mathbf{k}}}{2}\right) + \frac{1}{2}H^z \right) - \frac{1}{2}H^x A_{\eta=-1}^{z,mn}, \quad (22)$$

$$\begin{aligned} \tilde{H}^x C_{\mathbf{k}}^{+,mn} - \tilde{H}^x C_{\mathbf{k}}^{-,mn} &= \frac{1}{2}\tilde{H}^x (A_{\eta=-1}^{-,mn} - A_{\eta=-1}^{+,mn}) \\ &\quad - \epsilon_{\mathbf{k}} \coth\left(\frac{\beta\epsilon_{\mathbf{k}}}{2}\right) A_{\eta=-1}^{z,mn}. \end{aligned} \quad (23)$$

Because the observable correlations are in real space, we have to perform a corresponding Fourier transformation  $\langle (S^z)_i^m (S^-)_i^n S_i^{\alpha} \rangle = C_i^{\alpha,mn} = \frac{1}{N} \sum_{\mathbf{k}} C_{\mathbf{k}}^{\alpha,mn}$ .

Fourier transform of equation (22) yields

$$\begin{aligned} -C_i^{-,mn} + \frac{1}{N} \sum_{\mathbf{k}} \frac{H^x}{H^z} C_{\mathbf{k}}^{z,mn} &= \frac{1}{2} A_{\eta=-1}^{-,mn} \\ + \frac{1}{2} A_{\eta=-1}^{-,mn} \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{H^z} \coth\left(\frac{\beta\epsilon_{\mathbf{k}}}{2}\right) &- \frac{1}{2} A_{\eta=-1}^{z,mn} \frac{1}{N} \sum_{\mathbf{k}} \frac{H^x}{H^z}. \end{aligned} \quad (24)$$

Putting this into the Fourier transform of equation (21), one can eliminate the term  $\frac{1}{N} \sum_{\mathbf{k}} \frac{H^x}{H^z} C_{\mathbf{k}}^{z,mn}$ . This will turn out to be important in the later discussion of the eigenvector method where the formalism has to be modified because one cannot take the  $\mathbf{k}$ -dependent terms outside the sum (integral). One obtains

$$\begin{aligned} C_i^{+,mn} - C_i^{-,mn} - \frac{1}{2}(A_{\eta=-1}^{-,mn} - A_{\eta=-1}^{+,mn}) &= \\ \frac{1}{2}(A_{\eta=-1}^{-,mn} + A_{\eta=-1}^{+,mn}) \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{H^z} \coth\left(\frac{\beta\epsilon_{\mathbf{k}}}{2}\right). \end{aligned} \quad (25)$$

The Fourier transform of equation (23) can be done directly and gives

$$\begin{aligned} C_i^{+,mn} - C_i^{-,mn} - \frac{1}{2}(A_{\eta=-1}^{-,mn} - A_{\eta=-1}^{+,mn}) &= \\ - A_{\eta=-1}^{z,mn} \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{\tilde{H}^x} \coth\left(\frac{\beta\epsilon_{\mathbf{k}}}{2}\right). \end{aligned} \quad (26)$$

Equations (25), (26) are sufficient to determine the observables.

To elucidate these equations we derive the explicit expressions for spins  $S = 1/2$  and  $S = 1$ .

For  $S = 1/2$  we need  $m = 0$  and  $n = 1$ . This gives two equations of motion which determine  $\langle S^x \rangle$  and  $\langle S^z \rangle$ .

From equation (25) one finds with  $\langle S_i^- S_i^+ \rangle = 1/2 - \langle S_i^z \rangle$  and  $\langle S_i^- S_i^- \rangle = 0$  and  $A_{\eta=-1}^{01} = 0$  and  $A_{\eta=-1}^{+,10} = 2\langle S_i^z \rangle$

$$\frac{1}{2} = \langle S^z \rangle \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{H^z} \coth\left(\frac{\beta\epsilon_{\mathbf{k}}}{2}\right), \quad (27)$$

and from equation (26) with  $A_{\eta=-1}^{z,10} = -\langle S_i^- \rangle = -\langle S_i^x \rangle$

$$\frac{1}{2} = \langle S^x \rangle \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{\tilde{H}^x} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right). \quad (28)$$

These are two equations which determine the two unknowns  $\langle S^x \rangle$  and  $\langle S^z \rangle$ . No regularity conditions are necessary.

For  $S = 1$  one needs equations (25, 26) not only for  $(n = 1, m = 0)$  but also for  $(n = 1, m = 1), (n = 2, m = 0), (n = 3, m = 0)$ . This yields 8 equations for the eight unknowns  $\langle S^- \rangle, \langle S^z \rangle, \langle S^- S^- \rangle, \langle S^z S^- \rangle, \langle S^z S^z \rangle, \langle S^z S^z S^- \rangle, \langle S^z S^- S^- \rangle, \langle S^- S^- S^z S^z \rangle$ .

The left sides of equations (25) and (26) are the same. Therefore we write them pairwise. For  $(n = 1, m = 0)$  we have

$$2 - \langle S^z S^z \rangle - \langle S^- S^- \rangle = \begin{cases} \langle S^z \rangle \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{\tilde{H}^z} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right) \\ \langle S^- \rangle \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{\tilde{H}^x} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right). \end{cases} \quad (29)$$

For  $(n = 1, m = 1)$  and  $\langle S^z S^z S^z \rangle = \langle S^z \rangle$ , valid for  $S = 1$ , we have

$$\frac{1}{2} (\langle S^z + \langle S^z S^z \rangle - \langle S^- S^- \rangle - 2 \langle S^z S^- S^- \rangle - 2) = \begin{cases} \frac{1}{2} (\langle S^- S^- \rangle + 3 \langle S^z S^z \rangle - \langle S^z \rangle - 2) \\ \quad \times \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{\tilde{H}^z} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right) \\ \langle S^z S^- \rangle \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{\tilde{H}^x} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right). \end{cases} \quad (30)$$

For  $(n = 2, m = 0)$  and  $\langle S^- S^- S^- \rangle = 0$  we find

$$3 \langle S^- \rangle - \langle S^- S^z \rangle - \langle S^- S^z S^z \rangle + 2 \langle S^z S^- \rangle = \begin{cases} (2 \langle S^z S^- \rangle + \langle S^- \rangle) \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{\tilde{H}^z} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right) \\ 2 \langle S^- S^- \rangle \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{\tilde{H}^x} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right). \end{cases} \quad (31)$$

For  $(n = 3, m = 0)$  we have with  $\langle S^- S^- S^- S^- \rangle = 0$  and  $\langle S^- S^- S^- \rangle = 0$

$$2 \langle S^- S^- \rangle - \langle S^- S^- S^z \rangle - \langle S^- S^- S^z S^z \rangle = \begin{cases} (3 \langle S^z S^- S^- \rangle + 3 \langle S^- S^- \rangle) \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{\tilde{H}^z} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right) \\ 0. \end{cases} \quad (32)$$

From equations (32) we find  $\langle S^- S^- S^z S^z \rangle = 2 \langle S^- S^- \rangle - \langle S^- S^- S^z \rangle$  and  $\langle S^z S^- S^- \rangle = -\langle S^- S^- \rangle$ . The remaining correlations are determined by the previous six equations.

Instead of using these 8 equations one can also apply the regularity conditions, as we did in reference [5] when working with the commutator Green's functions. Then one can express all correlations in terms of the correlations  $\langle S^z \rangle$  and  $\langle S^z S^z \rangle$  in the case of spin  $S = 1$ , and in

terms of the moments  $\langle (S^z)^n \rangle$  with  $(n = 1, \dots, 2S)$  for arbitrary spin  $S$ , and one need only solve two equations in the  $S = 1$  case or  $2S$  equations for arbitrary spin values  $S$ .

The regularity conditions are obtained from the fact that the commutator Green's function has to be regular at the origin

$$\lim_{\omega \rightarrow 0} \omega G_{\mathbf{k}, \eta=-1}^{\alpha, mn} = 0, \quad (33)$$

which leads to the relations

$$\tilde{H}^x A_{\eta=-1}^{+, mn} + \tilde{H}^x A_{\eta=-1}^{-, mn} + 2H^z A_{\eta=-1}^{z, mn} = 0. \quad (34)$$

Note that these relations are also obtained by equating equations (25, 26).

For  $m = 0, n = 1$  we obtain the first regularity condition

$$\langle S^x \rangle = \frac{\tilde{H}^x}{H^z} \langle S^z \rangle; \quad (35)$$

*i.e.* the knowledge of  $\langle S^z \rangle$  determines  $\langle S^x \rangle$ .

From the definitions (12) one sees that the prefactor is momentum independent and the relation generalized to the multilayer can be written as

$$\langle S_i^x \rangle = \frac{\tilde{H}_i^x}{H_i^z} \langle S_i^z \rangle = \frac{B_i^x + J_{i,i+1} \langle S_{i+1}^x \rangle + J_{i,i-1} \langle S_{i-1}^x \rangle}{Z_i} \langle S_i^z \rangle. \quad (36)$$

In the case of spin  $S = 1$ , equation (25) with  $(n = 1, m = 0)$  and  $(n = 1, m = 1)$  together with the regularity conditions determines all desired correlations for spin  $S = 1$ . We demonstrate this by deriving the equations for  $\langle S^z \rangle$  and  $\langle S^z S^z \rangle$  already derived with the commutator Green's functions in reference [5].

Equation (25) gives for  $(n = 1, m = 0)$  with  $\langle S^- S^+ \rangle = 2 - \langle S^z \rangle - \langle S^z S^z \rangle$

$$2 - \langle S^z S^z \rangle - \langle S^- S^- \rangle = \langle S^z \rangle \frac{1}{N} \sum_{\mathbf{k}} \sqrt{1 + \frac{\tilde{H}^x H^x}{H^z H^z}} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right). \quad (37)$$

From the regularity conditions (34) one finds for  $S=1$

$$\langle S^- S^- \rangle = \frac{\left( \frac{\tilde{H}^x}{H^z} \right)^2}{2 - \left( \frac{\tilde{H}^x}{H^z} \right)^2} (3 \langle S^z S^z \rangle - 2). \quad (38)$$

Putting this into equation (37) gives the first of the desired equations

$$4 - 2 \langle S^z S^z \rangle \left( 1 + \left( \frac{\tilde{H}^x}{H^z} \right)^2 \right) - \langle S^z \rangle \left( 2 - \left( \frac{\tilde{H}^x}{H^z} \right)^2 \right) \times \frac{1}{N} \sum_{\mathbf{k}} \sqrt{1 + \frac{\tilde{H}^x H^x}{H^z H^z}} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right) = 0. \quad (39)$$

The second equation is obtained from equation (25) with  $(n = 1, m = 1)$ , and the regularity conditions which relates

$\langle S^z S^- S^- \rangle$  to  $\langle S^z \rangle$  and  $\langle S^z S^z \rangle$ . This leads to

$$\langle S^z \rangle \left( 2 - \left( \frac{\tilde{H}^x}{H^z} \right)^2 \right) - 2 \left( 3 \langle S^z S^z \rangle - 2 \right) \frac{1}{N} \sum_{\mathbf{k}} \sqrt{1 + \frac{\tilde{H}^x H^x}{H^z H^z}} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right) = 0. \quad (40)$$

The only difference from the corresponding equations of reference [5] is that the square root cannot be taken out of the sum (integral over the first Brillouin zone) because of the momentum dependence of  $H^x$  coming from the exchange anisotropy. If the latter is zero as in reference [5], only the single-ion anisotropy survives and one recovers the original equations.

The explicit derivations above are done for pedagogical reasons, but they would not have been necessary because one obtains by subtracting equation (21) from equation (22)

$$2\tilde{H}^x H^x C_{\mathbf{k}}^{z,mn} - \tilde{H}^x H^z C_{\mathbf{k}}^{+,mn} - \tilde{H}^x H^z C_{\mathbf{k}}^{-,mn} = \frac{1}{2} (A_{\eta=-1}^{-,mn} - A_{\eta=-1}^{+,mn}) \epsilon_{\mathbf{k}} \tilde{H}^x \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right) + \frac{1}{2} (A_{\eta=-1}^{+,mn} + A_{\eta=-1}^{-,mn}) \epsilon_{\mathbf{k}}^2 \frac{\tilde{H}^x}{H^z}, \quad (41)$$

which corresponds to equation (27) of reference [5] which is the starting point for deriving the equations for the moments explicitly.

For the treatment of multilayers we have to use the eigenvector method as mentioned above. The essential features are as follows. One starts with a transformation, which diagonalizes the  $\Gamma$ -matrix of equation (8)

$$\mathbf{L}\mathbf{\Gamma}\mathbf{R} = \mathbf{\Omega}, \quad (42)$$

where  $\mathbf{\Omega}$  is a diagonal matrix with eigenvalues  $\omega_{\tau}$  ( $\tau = 1, \dots, 3N$ ), and the transformation matrix  $\mathbf{R}$  and its inverse  $\mathbf{R}^{-1} = \mathbf{L}$  are obtained from the right eigenvectors of  $\mathbf{\Gamma}$  as columns and from the left eigenvectors as rows, respectively. These matrices are normalized to unity:  $\mathbf{R}\mathbf{L} = \mathbf{L}\mathbf{R} = \mathbf{1}$ .

Multiplying the equation of motion (8) from the left by  $\mathbf{L}$  and inserting  $\mathbf{1} = \mathbf{R}\mathbf{L}$  one finds

$$(\omega \mathbf{1} - \mathbf{\Omega}) \mathbf{L} \mathbf{G}_{\eta}^{mn} = \mathbf{L} \mathbf{A}_{\eta}^{mn}. \quad (43)$$

Defining  $\mathcal{G}_{\eta}^{mn} = \mathbf{L} \mathbf{G}_{\eta}^{mn}$  and  $\mathcal{A}_{\eta}^{mn} = \mathbf{L} \mathbf{A}_{\eta}^{mn}$  one obtains

$$(\omega \mathbf{1} - \mathbf{\Omega}) \mathcal{G}_{\eta}^{mn} = \mathcal{A}_{\eta}^{mn}. \quad (44)$$

$\mathcal{G}_{\eta}^{mn}$  is a new vector of Green's functions, each component  $\tau$  of which has but a single pole

$$\mathcal{G}_{\eta}^{mn,\tau} = \frac{\mathcal{A}_{\eta}^{mn,\tau}}{\omega - \omega_{\tau}}. \quad (45)$$

This is the important point and allows application of the spectral theorem to each component separately. This gives with  $\mathbf{C}^{mn} = \mathbf{L} \mathbf{C}^{mn}$

$$\mathcal{C}^{mn,\tau} = \frac{\mathcal{A}_{\eta}^{mn,\tau}}{e^{\beta \omega_{\tau}} + \eta} + \frac{1}{2} (1 - \eta) \frac{1}{2} \lim_{\omega \rightarrow 0} \frac{\omega \mathcal{A}_{\eta}^{mn,\tau}}{\omega - \omega_{\tau}}. \quad (46)$$

In reference [9] we used the commutator ( $\eta = -1$ ). Here, we proceed with the anticommutator ( $\eta = +1$ ), so that the second term in equation (46) is zero and one obtains the original correlation vector  $\mathbf{C}^{mn}$  by multiplying  $\mathcal{C}^{mn}$  from the left with  $\mathbf{R}$ ; *i.e.*

$$\mathbf{C}^{mn} = \mathbf{R} \mathcal{E} \mathbf{L} \mathbf{A}_{\eta=+1}^{mn}, \quad (47)$$

where  $\mathcal{E}$  is a diagonal matrix with matrix elements  $\mathcal{E}_{ij} = \delta_{ij} (e^{\beta \omega_i} + 1)^{-1}$ . With the relation (20) we find

$$\mathbf{C}^{mn} = \mathbf{R} \mathcal{E} \mathbf{L} (\mathbf{A}_{\eta=-1}^{mn} + 2\mathbf{C}^{mn}), \quad (48)$$

or

$$\mathbf{C}^{mn} = (\mathbf{1} - 2\mathbf{R} \mathcal{E} \mathbf{L})^{-1} \mathbf{R} \mathcal{E} \mathbf{L} \mathbf{A}_{\eta=-1}^{mn}. \quad (49)$$

For the monolayer it can be shown explicitly that the eigenvector method yields the equations (21, 22, 23) derived before. In this case the eigenvectors by which the transformation matrices  $\mathbf{R}$  and  $\mathbf{L}$  are constructed can be given explicitly. They are

$$\mathbf{R} = \begin{pmatrix} \frac{H^x}{H^z} & \frac{-(\epsilon_{\mathbf{k}} + H^z)}{H^x} & \frac{(\epsilon_{\mathbf{k}} - H^z)}{H^x} \\ \frac{H^x}{H^z} & \frac{(\epsilon_{\mathbf{k}} - H^z)}{H^x} & \frac{-(\epsilon_{\mathbf{k}} + H^z)}{H^x} \\ 1 & 1 & 1 \end{pmatrix}, \quad (50)$$

and

$$\mathbf{L} = \frac{1}{4\epsilon_{\mathbf{k}}^2} \begin{pmatrix} 2\tilde{H}^x H^z & 2\tilde{H}^x H^z & 4H^z H^z \\ -(\epsilon_{\mathbf{k}} + H^z) \tilde{H}^x & (\epsilon_{\mathbf{k}} - H^z) \tilde{H}^x & 2H^x \tilde{H}^x \\ (\epsilon_{\mathbf{k}} - H^z) \tilde{H}^x & -(\epsilon_{\mathbf{k}} + H^z) \tilde{H}^x & 2H^x \tilde{H}^x \end{pmatrix}. \quad (51)$$

Putting this into equation (48) yields equations (21, 22, 23).

In order to obtain the correlations in real space, equation (49) has to be Fourier transformed and the resulting integral equation has to be solved self-consistently. By inspecting the expressions for the monolayer, one can show that the inverse  $(\mathbf{1} - 2\mathbf{R} \mathcal{E} \mathbf{L})^{-1}$  does not exist. Therefore this equation cannot furnish the solution. However, one can show that the formulation with the anticommutator relation can be transformed into the result for the commutator relation:  $\mathcal{E}$  is a diagonal matrix with  $\mathcal{E}_{ij} = \delta_{ij} (e^{\beta \omega_i} + 1)^{-1}$ . With the relation  $(\mathbf{R} \mathcal{E} \mathbf{L})^{-1} = \mathbf{L}^{-1} (\mathcal{E})^{-1} \mathbf{R}^{-1} = \mathbf{R} \mathcal{E}^{-1} \mathbf{L}$  one obtains from equation (49)

$$\begin{aligned} \mathbf{C} &= (\mathbf{R} (\mathbf{1} - 2\mathcal{E}) \mathbf{L})^{-1} \mathbf{R} \mathcal{E} \mathbf{L} \mathbf{A} \\ &= \mathbf{R} (\mathbf{1} - 2\mathcal{E})^{-1} \mathcal{E} \mathbf{L} \mathbf{A} \\ &= \mathbf{R} \tilde{\mathcal{E}} \mathbf{L} \mathbf{A}, \end{aligned} \quad (52)$$

where  $\tilde{\mathcal{E}}_{ij} = \delta_{ij} \frac{\mathcal{E}_{ii}}{1 - 2\mathcal{E}_{ii}} = \delta_{ij} (e^{\beta \omega_i} - 1)^{-1}$ , which still is of no use because it diverges for  $\omega_i = 0$ .

In reference [9], it was shown that the matrix  $\mathbf{R}_0\mathbf{L}_0$ , where the index refers to the eigenvectors with eigenvalue zero, is a projection operator onto the zero eigenvalue space. The situation can then be remedied by projecting equation (52) onto the non-zero eigenvalue space with  $\mathbf{1} - \mathbf{R}_0\mathbf{L}_0$ , which leads to the commutator expression for the correlations of reference [9]

$$\mathbf{C} = \mathbf{R}\tilde{\mathcal{E}}_0\mathbf{L}\mathbf{A} + \mathbf{R}_0\mathbf{L}_0\mathbf{C}, \quad (53)$$

where  $\tilde{\mathcal{E}}_0$  is equal to  $\tilde{\mathcal{E}}$  in which the diagonal elements corresponding to  $\omega = 0$  have been set to zero.

The problem one is confronted with in applying the eigenvector method to this equation is that the exchange anisotropy introduces a momentum dependence in the projection operator. Then the projector cannot be taken out of the integral when a Fourier transform to real space is performed as in the case of the Anderson Callen decoupling of the single-ion anisotropy only. The way out is to eliminate the projector by a transformation in one component of equation (53), which is sufficient to establish the integral equations of the eigenvector method. This procedure was inspired by the elimination of the disturbing term in equations (21, 22).

The adequate transformation is found to be

$$\mathbf{T}^{-1} = \frac{1}{2} \begin{pmatrix} 1 & 1 & 0 \\ -1 & 1 & 0 \\ 0 & 0 & 2 \end{pmatrix} \quad \mathbf{T} = \begin{pmatrix} 1 & -1 & 0 \\ 1 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (54)$$

with  $\mathbf{T}^{-1}\mathbf{T} = \mathbf{1}$ .

Applying this transformation to equation (53)

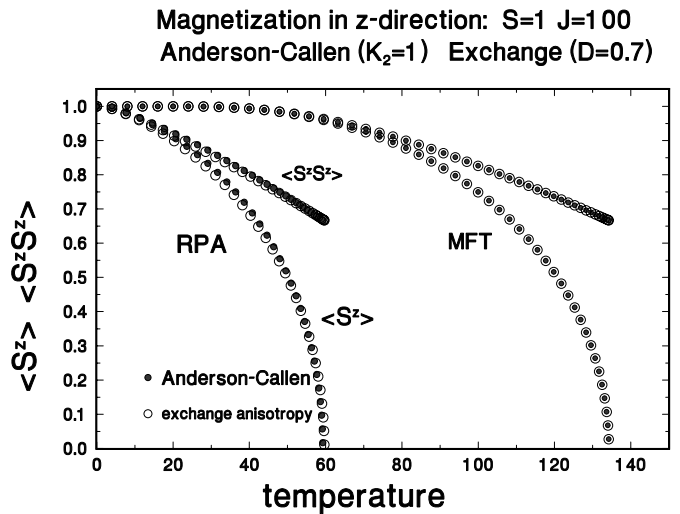
$$\mathbf{T}^{-1}\mathbf{C} = \mathbf{T}^{-1}\mathbf{R}\tilde{\mathcal{E}}_0\mathbf{L}\mathbf{T}\mathbf{T}^{-1}\mathbf{A} + \mathbf{T}^{-1}\mathbf{R}_0\mathbf{L}_0\mathbf{T}\mathbf{T}^{-1}\mathbf{C} \quad (55)$$

and inserting the eigenvectors (50,51) transforms the second component of the vector  $\mathbf{T}^{-1}\mathbf{R}_0\mathbf{L}_0\mathbf{T}\mathbf{T}^{-1}\mathbf{C}$  to zero, and one recovers equation (25) from the second row of the transformed equation (55), from which, together with the regularity conditions, the integral equations for the correlations for each  $(m, n)$ -pair are obtained.

The eigenvector method is then immediately generalized to the case of  $N$  layers by applying the transformation to equation (53) read as a  $3N$ -dimensional problem, thus constructing  $3N \times 3N$ -matrices with sub-matrices formed with the transformation (54) on the diagonals.

### 3 Numerical results

In this section we discuss results of calculations for a square lattice including the exchange anisotropy. The integral equations (55) for determining the correlations together with the regularity conditions as derived in Appendix A of this paper are solved self-consistently by the curve following method, which we described in detail in Appendix A of reference [7]. A comparison with the results for the single-ion anisotropy as used in our previous work [5,9] is effected by fixing the strength of the

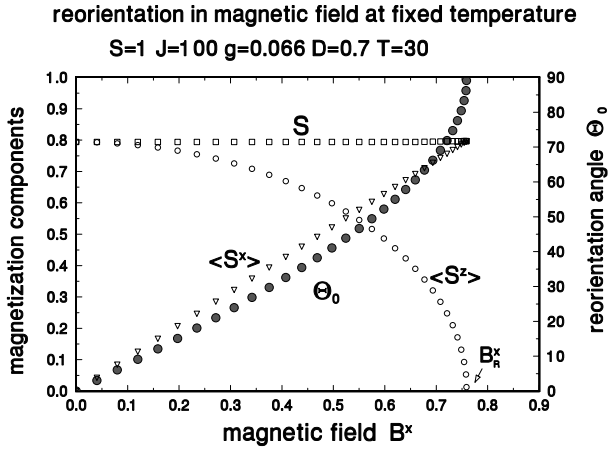


**Fig. 1.** The magnetization  $\langle S^z \rangle$  and  $\langle S^z S^z \rangle$  of a ferromagnetic spin  $S = 1$  Heisenberg monolayer for a square lattice are shown as functions of the temperature (no magnetic field). Comparison is made between Green's function (RPA) calculations using the exchange anisotropy ( $D = 0.7$ , open circles) and the single-ion anisotropy ( $K_2 = 1$ , solid dots) with Anderson-Callen decoupling. The corresponding results of mean field (MFT) calculations are also shown.

exchange anisotropy such that the Curie temperature of a square monolayer with spin  $S = 1$  agrees approximately with that of a single-ion anisotropy with strength  $K_2 = 1$ , which was used in most of the calculations of our previous work. The exchange interaction strength,  $J = 100$ , and the dipole coupling strength,  $g = 0.018$  (corresponding to the case of Ni), are taken to be the same in both kinds of calculations. The exchange anisotropy coupling strength turns out to be  $D = 0.7$ . In order to compare results for different spin values all parameters are scaled as  $J \rightarrow J/S(S + 1)$ ,  $g \rightarrow g/S(S + 1)$ ,  $D \rightarrow D/S(S + 1)$ ,  $K_2 \rightarrow K_2/S(S - 1/2)$  and if a magnetic field is applied,  $\mathbf{B} \rightarrow \mathbf{B}/S$ .

In Figure 1 we display the magnetization  $\langle S^z \rangle$  and its second moment  $\langle S^z S^z \rangle$  as functions of the temperature for a  $S = 1$  monolayer using Green's function theory (denoted as RPA). Using a value of  $D = 0.7$  as the strength of the exchange anisotropy yields nearly the same curve as for an Anderson-Callen decoupling of the single-ion anisotropy with a strength of  $K_2 = 1$ . The results for the corresponding mean field (MFT) calculations also practically coincide with each other. As is well known and also discussed in our previous work, the neglect of magnons results in a Curie temperature which is more than a factor of two larger than that obtained by including magnon excitations. This difference for a monolayer is much larger than the corresponding difference for bulk ferromagnets.

In Figure 2 we show the reorientation of the magnetization at a fixed temperature due to a transverse field in  $x$ -direction,  $B^x$ . The magnetic field in  $z$ -direction is set to zero,  $B^z = 0$ . In this case, the strength of the dipole coupling is chosen to be  $g = 0.066$  (a value corresponding to Co). As a function of the external field,

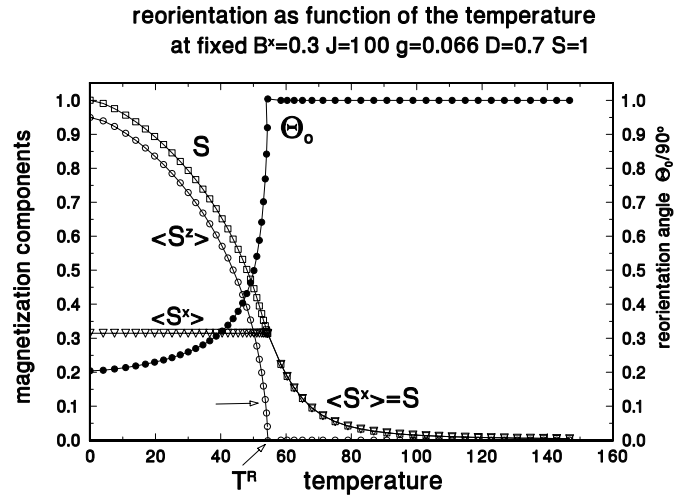


**Fig. 2.** The components of the magnetization  $\langle S^z \rangle$  and  $\langle S^x \rangle$  and the absolute value  $S$  at a fixed temperature  $T = 30$  as function of an external magnetic field in the  $x$ -direction,  $B^x$ , are shown for a ferromagnetic spin  $S = 1$  Heisenberg monolayer for a square lattice. Also shown are the equilibrium reorientation angle  $\theta_0$  and the critical reorientation field,  $B_R^x$ , at which in-plane orientation is reached.

the  $x$ -component of the magnetization  $\langle S^x \rangle$  rises linearly, whereas its  $z$ -component  $\langle S^z \rangle$  falls to zero, where the in-plane magnetization is reached ( $\theta_0 = 90^\circ$ ). The absolute value  $S = \sqrt{\langle S^x \rangle^2 + \langle S^z \rangle^2}$  remains constant, as it should do, and is also shown in the figure.

In Figure 3, we show the magnetization components as functions of the temperature for the same parameters as in Figure 2 at a constant external field,  $B^x = 0.3$ . The component  $\langle S^x \rangle$  stays constant until the component  $\langle S^z \rangle$  has dropped to zero, and an in-plane magnetization ( $\theta_0 = 90^\circ$ ) is reached. The fact that  $\langle S^x \rangle$  is constant for temperatures below the reorientation temperature,  $T < T_R$ , can be understood from the regularity condition (35), which, for a monolayer with scaled parameters ( $\langle S^z \rangle$  drops out of the expression if  $B^z = 0$ ), is  $\langle S^x \rangle / S = \frac{B^x / S}{(4D - 3gT^0) / S(S+1)}$ . This expression restricts the range of parameters for the validity of our approach because  $\langle S^x \rangle / S$  must be  $\leq 1$ . Above the reorientation temperature,  $\langle S^z \rangle$  remains zero whereas  $\langle S^x \rangle$  remains finite because of the field in the  $x$ -direction and decreases slowly with increasing temperature. The absolute value of the magnetization  $S$  is also shown; above  $T_R$  one has  $S = \langle S^x \rangle$ .

With the chosen parameters one observes a novel feature in the calculations which is connected with the introduction of the exchange anisotropy *together* with the dipole coupling. In this case, the eigenvalues of equation (17) become complex above a certain temperature, *i.e.* below a certain value of  $\langle S^z \rangle$ . This behaviour occurs quite naturally in the theory. Because the  $\Gamma$ -matrix of equation (8) is real, its eigenvalues and eigenvectors, if complex, occur pairwise as complex conjugates, and the term  $\mathbf{R}\mathcal{E}\mathbf{L}$  in the equation (55) is real, so that one has to do with a real integral equation. The complex eigenvalues and vectors have to be taken seriously, and are necessary for obtaining the results of Figure 3. The complex eigen-



**Fig. 3.** The components of the magnetization  $\langle S^z \rangle$  and  $\langle S^x \rangle$  and its absolute value  $S$  for a fixed magnetic field  $B^x = 0.3$  as function of the temperature are shown for a ferromagnetic spin  $S = 1$  Heisenberg monolayer for a square lattice. Also shown are the equilibrium reorientation angle  $\theta_0$  and the critical reorientation temperature,  $T_R$ , at which in-plane orientation is reached. The small horizontal arrow indicates the value of  $\langle S^z \rangle$  below which complex eigenvalues occur.

values are connected with the additional term in  $H_i^\pm$  of equation (12), which comes from the exchange anisotropy. This can be seen analytically by considering the dispersion relation (17), which shows that the optimal condition for the occurrence of complex eigenvalues is at  $\mathbf{k} = 0$  (thus  $\gamma_{\mathbf{k}} = 4$ ). With  $q = 4$ ,  $B^z = 0$ ,  $K_2 = 0$  one has for the monolayer from equation (17)

$$\epsilon_{\mathbf{k}=0}^2 = (4D - 2gT^0)^2 \langle S^z \rangle^2 - (gT^0 \langle S^x \rangle + B^x) (\langle S^x \rangle (4D - gT^0) - B^x). \quad (56)$$

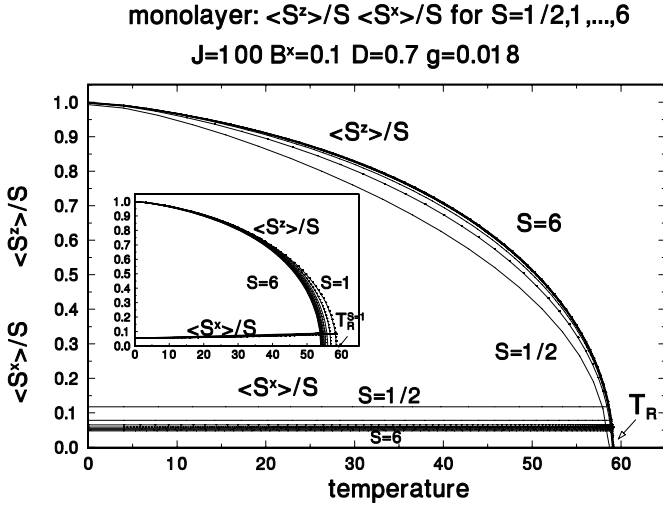
Complex eigenvalues occur if the second term is larger than the first term. Using the regularity condition (37) for  $\langle S^x \rangle$ , one obtains (using scaled parameters for  $S = 1$ ) a condition for the occurrence of complex eigenvalues,

$$\langle S^z \rangle < \frac{2B^x}{(4D - 3gT^0)} \sqrt{\frac{2gT^0}{(4D - 2gT^0)}}. \quad (57)$$

This yields complex eigenvalues for  $\langle S^z \rangle < 0.1157$  at  $T \simeq 53$  for the parameters used in Figure 3. This is confirmed in the numerical calculations, where of course complex eigenvectors also occur at finite  $\mathbf{k}$ . Enlarging  $B^x$  and/or  $g$  increases the range of complex eigenvalues. No complex eigenvalues occur if the dipole coupling is set equal to zero. In our previous work [9], where we used the single-ion anisotropy together with the dipole coupling, complex eigenvalues never occurred, which can be understood from the structure of the dispersion relation (17) by putting the exchange anisotropy terms equal to zero.

In Figure 4 we display the normalized magnetizations  $\langle S^z \rangle / S$  of a monolayer as functions of the temperature  $T$  for all half-integral and integral spin values ranging from

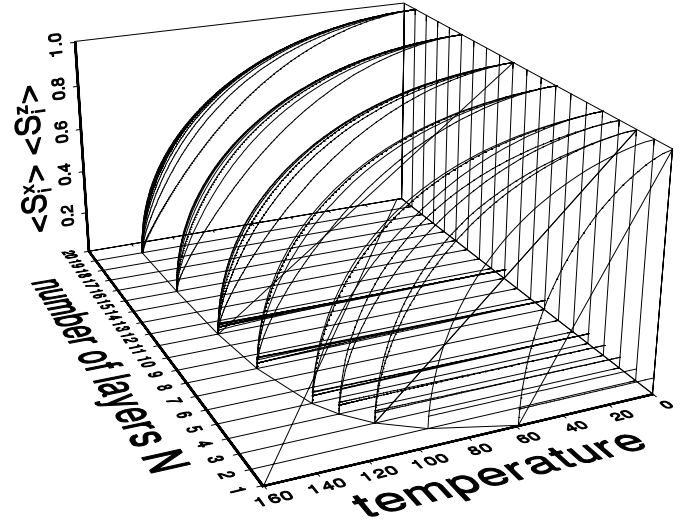




**Fig. 4.** Normalized magnetization curves for  $\langle S^z \rangle / S$  and  $\langle S^x \rangle / S$  for a monolayer with spin values between  $S = 1/2$  and  $S = 6$  shown as functions of the temperature using the exchange anisotropy. The corresponding results obtained with a single-ion anisotropy ( $K_2 = 1$ ) are shown in the inset (from Ref. [9]).

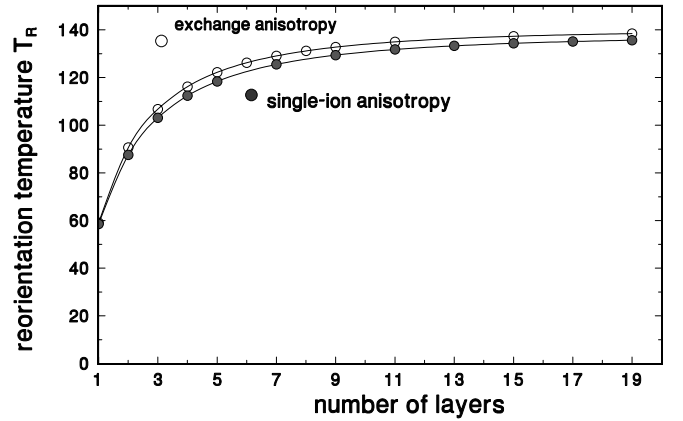
$S = 1/2$  to  $S = 6$ . In the case of the exchange anisotropy, the reorientation temperature  $T_R$  is practically the same for all spin values. One observes a spin dependence of the magnetization curves which decreases with increasing spin. The curves, however, saturate very quickly; on the scale of the figure, one cannot distinguish the curves between  $S = 2$  and  $S = 6$ . When using the single-ion anisotropy (the results are shown as an inset) there is a difference in the reorientation temperatures, but the magnetization curves again saturate very quickly (approaching the classical limit) but in the opposite direction. In both cases, the values for  $\langle S^x \rangle / S$  remain rather small owing to the application of the small field in the  $x$ -direction  $B^x = 0.1$ .

In Figure 5, we display the sublayer magnetization components  $\langle S_i^z \rangle$  and  $\langle S_i^x \rangle$  as functions of the temperature for spin  $S = 1$  films with thicknesses ranging from  $N = 1$  to  $N = 19$  layers calculated with the exchange anisotropy using the same parameters as in Figure 4. The reorientation temperatures  $T_R^N$  can be read from the curve in the  $N$ - $T$  plane, which approaches the bulk value with increasing layer thickness. Similar results were obtained in reference [9] when using the single-ion anisotropy. To have a direct comparison of the reorientation temperatures calculated with the exchange anisotropy ( $D = 0.7$ ) and the single-ion anisotropy ( $K_2 = 1$ ), we display Figure 6. Because the exchange anisotropy parameter was fitted to give the same result for the Curie temperature as a calculation with the anisotropy parameter for the monolayer, the corresponding reorientation temperatures practically coincide in this case, whereas the reorientation temperatures turn out to be slightly higher for the exchange anisotropy calculations with increasing film thickness. The satura-



**Fig. 5.** Sublayer magnetization components  $\langle S_i^z \rangle$  and  $\langle S_i^x \rangle$  as functions of the temperature for spin  $S = 1$  films with  $N$  layers calculated with the exchange anisotropy. The reorientation temperatures  $T_R^N$  can be read from the curve in the  $N$ - $T$  plane.

#### reorientation temperature versus film thickness



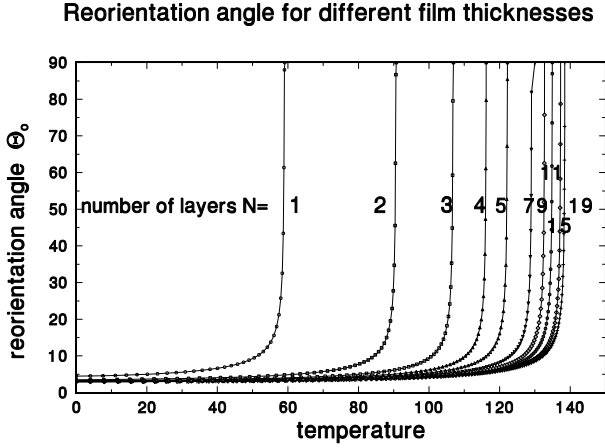
**Fig. 6.** Reorientation temperature as a function of the film thickness displayed for results with the exchange anisotropy and the Anderson-Callen treatment of the single-ion anisotropy (see Ref. [9] for the latter).

tion towards the bulk limit follows the same trend in both cases.

In Figure 7 we show the average reorientation angle  $\theta_0(N, T)$  as a function of the temperature for films with increasing film thickness, where we define

$$\theta_0(N, T) = \arctan \frac{\frac{1}{N} \sum_{i=1}^N \langle S_i^z \rangle}{\frac{1}{N} \sum_{i=1}^N \langle S_i^x \rangle}. \quad (58)$$

The curves show the same saturation behaviour in the bulk limit as already seen in the previous figures.



**Fig. 7.** Average equilibrium reorientation angle for different film thicknesses (number of layers  $N = 1, \dots, 5, 7, 9, 11, 15, 19$ ) shown as a function of the temperature. This is a result of calculations using the exchange anisotropy and the parameters are the same as in the previous figures.

## 4 Conclusions

In the present paper, we have included the exchange anisotropy in our many-body Green's function description of thin ferromagnetic films. A few non-trivial changes in the general formalism had to be implemented because of additional momentum dependencies when the exchange anisotropy is included. The momentum dependence of the projector onto the zero eigenvalue space has been eliminated by an appropriate transformation. A novel feature is the appearance of complex eigenvalues and eigenvectors of the non-symmetric matrix governing the equation of motion for the Green's functions when including the exchange anisotropy and the dipole coupling together. The complex eigenvalues occur quite naturally in the theory and have to be taken seriously. They are necessary to obtain correct results above the temperature where the magnetization  $\langle S^z \rangle$  drops below a certain value.

The physical quantities calculated with the exchange anisotropy or with the single-ion anisotropy are fairly similar when the exchange anisotropy strength is fitted in such a way that it gives the same Curie temperature for a spin  $S = 1$  Heisenberg monolayer as that calculated with the single-ion anisotropy parameter of a certain strength, the rest of the parameters such as the exchange interaction strength and the dipole coupling strength being the same.

In the present paper, all intra- and interlayer coupling parameters have been taken to be the same but the computer program is written in such a way that they can easily be chosen differently. We have also shown only examples for a monolayer with spins in the range between  $S = 1$  and  $S = 6$  and films with  $N$  layers for spin  $S = 1$ . It is only a question of computer time to make calculations for films with other  $(S, N)$  combinations.

We are indebted to P.J. Jensen for very useful discussions.

## Appendix A: Treating $S \geq 1$

In this Appendix we show how the regularity conditions can be deduced for general spin quantum numbers  $S$  and for multilayers. From the definitions (12) we see from (35) that

$$\frac{\tilde{H}_i^x}{H_i^z} = \frac{B_i^x}{Z_i}. \quad (59)$$

The regularity conditions (34) can therefore be written for general  $m, n$  in the form for each layer  $i$

$$-2Z_i A_{-1,i}^{z,mn} = A_{-1,i}^{+,mn} B_i^x + A_{-1,i}^{-,mn} B_i^x. \quad (60)$$

For the calculation of the correlations for higher spin we use equation (25) generalized to the multilayer case. We leave out the layer index  $i$  in all formulas which follow.

$$\begin{aligned} & \langle (S^z)^m (S^-)^n S^+ \rangle - \langle (S^z)^m (S^-)^n S^- \rangle \\ & - \frac{1}{2} (A_{\eta=-1}^{-,mn} - A_{\eta=-1}^{+,mn}) = \frac{1}{2} (A_{\eta=-1}^{-,mn} + A_{\eta=-1}^{+,mn}) \\ & \times \frac{1}{N} \sum_{\mathbf{k}} \frac{\epsilon_{\mathbf{k}}}{H^z} \coth \left( \frac{\beta \epsilon_{\mathbf{k}}}{2} \right). \quad (61) \end{aligned}$$

We express all correlation functions occurring in this equation in a standard form where all powers of  $S^z$  are written to the left of the powers of  $S^-$ :

$$C(m, n) = \langle (S^z)^m (S^-)^n \rangle. \quad (62)$$

Then, with the relations  $[S^z, (S^-)^n]_- = -n(S^-)^n$  and  $S^- S^+ = S(S+1) - S^z - (S^z)^2$ , we find that

$$\begin{aligned} & \langle (S^z)^m (S^-)^n S^z \rangle = nC(m, n) + C(m+1, n), \\ & \langle (S^z)^m (S^-)^n S^+ \rangle = \left( S(S+1) - n(n-1) \right) C(m, n-1) \\ & - (2n-1)C(m+1, n-1) \\ & - C(m+2, n-1), \\ & \langle (S^z)^m (S^-)^n S^- \rangle = C(m, n+1). \quad (63) \end{aligned}$$

The commutators can also be expressed in terms of the  $C(m, n)$  using the binomial series

$$\begin{aligned} & A_{-1}^{z,mn} = -nC(m, n), \\ & A_{-1}^{+,mn} = \left\langle \left[ \left( (S^z - 1)^m - (S^z)^m \right) S^- S^+ + 2S^z (S^z - 1)^m \right. \right. \\ & \quad \left. \left. + (n-1)(n+2S^z)(S^z)^m \right] (S^-)^{n-1} \right\rangle \\ & = S(S+1) \sum_{i=1}^m \binom{m}{i} (-1)^i C(m-i, n-1) \\ & \quad + (2n+m)C(m+1, n-1) \\ & \quad + \sum_{i=2}^{m+1} \binom{m+1}{i} (-1)^{i+1} C(m+2-i, n-1) \\ & \quad + n(n-1)C(m, n-1), \\ & A_{-1}^{-,mn} = \left\langle \left[ (S^z + 1)^m - (S^z)^m \right] (S^-)^{n+1} \right\rangle \\ & = \sum_{i=1}^m \binom{m}{i} C(m-i, n+1). \quad (64) \end{aligned}$$

Now by putting equation (64) into equation (60) the regularity conditions for all  $m$  and  $n$  can be written in terms of correlations defined in the standard form:

$$\begin{aligned}
2ZnC(m, n) = & B^x \left[ S(S+1) \sum_{i=1}^m \binom{m}{i} (-1)^i C(m-i, n-1) \right. \\
& + (2n+m)C(m+1, n-1) \\
& + \sum_{i=2}^{m+1} \binom{m+1}{i} (-1)^{i+1} C(m+2-i, n-1) \\
& \left. + n(n-1)C(m, n-1) \right] + B^x \sum_{i=1}^m \binom{m}{i} C(m-i, n+1).
\end{aligned} \tag{65}$$

For a given spin  $S$ , this set of linear equations for the correlations has to be solved for all  $m+n \leq 2S+1$ . The solutions have to be put *via* equations (63) together with (64) into equations (61), thus leading to a set of  $2S$  equations for the moments  $\langle (S^z)^p \rangle$  ( $p = 1, \dots, 2S$ ), which have to be solved self-consistently. The highest moment  $\langle (S^z)^{2S+1} \rangle$  has been eliminated in favour of the lower ones through the relation  $\prod_{M_S} (S^z - M_S) = 0$ .

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