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Magnetic correlations in ferromagnets undergoing spin reorientation transitions

A del Moral

Laboratorio de Magnetismo, Departamento de Física de Materia Condensada and Instituto de Ciencia de Materiales de Aragón (ICMA), Universidad de Zaragoza and Consejo Superior de Investigaciones Científicas (CSIC), 50009 Zaragoza, Spain

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Abstract. The longitudinal (parallel to the c axis) and transverse (to the c axis) spin correlation functions are calculated for ferromagnetic systems with crystalline axial symmetries, and undergoing spin reorientation (SR) transitions under the effects of competing crystal electric field (CEF) anisotropies of axial and planar character. The structure factor is a combination of a long-range ferromagnetic one, $F(\theta)\delta(k)$, with amplitude dependent on the SR angle θ , plus a finite contribution at finite wavevectors k . This small-angle neutron scattering (SANS) structure factor is contributed by a Lorentzian (L) term, due to the magnon scattering, plus a squared Lorentzian (L^2) term produced by zero-point uniform quantum spin fluctuations. This SANS scattering in the canted magnetization phase is produced within the magnetic domain walls, and therefore has a correlation length of the order of the domain wall width. The prefactors of the L and L^2 SANS factors are complex functions of the spin reorientation angle and of the CEF strength parameters and spin-wave stiffness constant. These results are briefly discussed in connection with recent SANS experiments performed in the tetragonal $(\text{Er}_x\text{Nd}_{1-x})_2\text{Fe}_{14}\text{B}$ hard intermetallic pseudoternary compounds.

1. Introduction

Pseudoternary rare-earth intermetallics, such as $(\text{Er}_x\text{RE}_{1-x})_2\text{Fe}_{14}\text{B}$ ($\text{RE} = \text{Nd}, \text{Dy}, \text{Ho}$) and $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$, are crystallographically uniaxial (tetragonal or hexagonal) ferromagnets. The presence of the RE ions with opposite α_j Stevens coefficients produces competing axial–planar magnetocrystalline anisotropies, which give rise to the appearance of a spin reorientation (SR) process. The overall magnetization vector rotates from the crystallographic c axis towards the basal plane, the system becoming a canted ferromagnet, the magnetization forming a temperature-dependent canting angle θ with the c axis (Ibarra *et al* 1988). This process starts below a certain phase transition temperature T_{SR} , around which the order parameter θ is believed to suffer strong critical fluctuations (del Moral *et al* 1989, 1990). Usually the transition-metal partner cooperates in getting the high-temperature axial magnetic structure. Sometimes, as is the case for the pure $\text{Nd}_2\text{Fe}_{14}\text{B}$ system, the competition between second- and fourth-order crystal electric field (CEF) anisotropies is responsible for the transition, although we are not going to treat this situation here. That this is a second-order phase transition (Algarabel *et al* 1988) has been demonstrated through the anomalies appearing at T_{SR} in the low-field AC magnetic susceptibility (Algarabel *et al* 1988, del Moral *et al* 1989, Ibarra *et al*

1989b) and also from the scaling of θ near T_{SR} (Moze *et al* 1990). More details about this transition and its critical character are given in the above references.

In a companion paper to this one, hereafter termed paper I (del Moral 1992a), a model, based on the spin-wave approximation, was developed. There, a mechanism was proposed to explain such a transition, as well as a calculation of the temperature dependence of the order parameter θ . In particular, this model determines the value of the critical exponent β ($=1/2$) and the low-temperature dependence of $\sin^2\theta$, of $T^{3/2}$ Bloch-like type. Also predicted is the relation between the 0 K SR angle $\theta(0)$ and T_{SR} . This angle is mainly due to the uniform ($k = 0$) zero-point quantum spin fluctuations.

On the other hand, small-angle neutron scattering (SANS) experiments performed on the whole $(Er_xNd_{1-x})_2Fe_{14}B$ series (del Moral *et al* 1991) showed a series of features. The structure factor is essentially of squared Lorentzian profile, with strongly dependent temperature amplitudes, which show anomalies at T_{SR} and T_{SR2} , the temperature where a cone-cone transition is believed to occur (del Moral *et al* 1989). The associated coherence lengths, of the order of ≈ 150 – 300 Å, show as well strong temperature dependences and weak, but well defined, anomalies in the form of peaks at T_{SR} . Therefore we have tried in this paper to develop a model, based also on the spin-wave approximation, in order to explain the existence of SANS scattering in a canted ferromagnet, as well as its origin. The model presented here is intimately connected with the one in paper I, and essentially deals with the calculation of the spin correlation functions, parallel and transverse to the c axis, together with the temperature dependences of the amplitude and correlation length associated with those factors. A very simplified and brief version of the model, only valid above and below but very close to T_{SR} , has already been reported (del Moral *et al* 1991). A quantitative comparison of our findings with SANS experiments in the $(Er_xNd_{1-x})_2Fe_{14}B$ series can be found elsewhere (del Moral *et al* 1992b).

2. Outline of the spin-wave model for spin reorientation transitions

The model developed for spin reorientation transitions in systems with competing axial-planar anisotropies was, as already mentioned, developed in the companion paper I. Therefore, I will only outline here the main features of the model in order to make the present paper more transparent and comprehensive, but restricting the results presented only to those relevant for the calculation of the spin correlation functions, which is the objective of the present work.

The complex alloys considered in section 1 are assumed to be well described by an effective magnetic moment $\mu = g\mu_B S$, where g and S respectively are the effective Landé factor and spin angular momentum, this moment being the one per lattice cell. A discussion and a justification of this procedure is given in paper I. The Hamiltonian describing our system is assumed to be

$$H = H_{ex} + H_a + H_p = -\frac{1}{2} \sum_{l,m} J(l-m) S_l \cdot S_m - D_a \sum_l \bar{O}_2^0(S_l) - D_p \sum_l \bar{O}_2^2(S_l) \quad (1)$$

where \bar{O}_2^0 and \bar{O}_2^2 are extended Stevens operators (Buckmaster 1962, Rudowicz 1985),

$$\bar{O}_2^0 = \frac{1}{2}[3S_z^2 - S(S+1)] \quad \bar{O}_2^2 = (\sqrt{6}/8)(S_+^2 + S_-^2) = (\sqrt{6}/8)[(S^x)^2 - (S^y)^2]. \quad (2)$$

In (1), H_{ex} represents the isotropic exchange interaction between S_l and S_m spins with

strength J , and H_a and H_p are competing, axial and planar respectively, crystal-field anisotropies, of strengths D_a and D_p , and responsible for the spin reorientation.

The spin-wave approximation that we are going to use naturally assumes small spin deviations from the average magnetization (M) axis; therefore, and in the presence of an SR of M , we need to take a frame with the new quantization axis (QA) $z' \parallel M$. Therefore, we need to rotate the crystal frame axes, with $OX \parallel a$, $OY \parallel b$ and $OZ \parallel c$ (where a , b and c are the crystal axes), by Eulerian angles ψ around OZ until the plane (c, M) , and θ around the new OY' axis until $OZ' \parallel M$. Then θ and ψ are the spherical coordinates of M in the crystal axes frame. Accordingly, we have to rotate Hamiltonian H in the way detailed in I.

The procedure of the transformation of the rotated Hamiltonian from (1) to the magnon representation is fully detailed in I, and here we will only give an outline of the procedure followed, finishing with the results that are relevant to the calculation of the spin correlation functions. Namely, in terms of creation, a_l^\dagger , and annihilation, a_l , spin deviation operators at site l , and for large spin S , we first write

$$S^z(l) = S - a_l^\dagger a_l \quad S^+(l) = \sqrt{(2S)}a_l \quad S^-(l) = \sqrt{(2S)}a_l^\dagger \quad (3)$$

which immediately gives

$$S^x(l) = [\sqrt{(2S)}/2](a_l + a_l^\dagger) \quad S^y(l) = [\sqrt{(2S)}/2](a_l - a_l^\dagger). \quad (4)$$

Afterwards we transform to the magnon representation, according to the standard Holstein-Primakoff (HP) transformations,

$$a_l^\dagger = \frac{1}{\sqrt{N}} \sum_k a_k^\dagger e^{ik \cdot l} \quad a_l = \frac{1}{\sqrt{N}} \sum_k a_k e^{-ik \cdot l}. \quad (5)$$

The rotated magnon Hamiltonian is

$$H' = H_0 + \sum_k (A_k a_k^\dagger a_k + \frac{1}{2} B_k a_k a_{-k} + \frac{1}{2} B_k^\dagger a_k^\dagger a_{-k}^\dagger) + (\rho_0 a_0 + \rho_0^\dagger a_0^\dagger) \quad (6)$$

where the form of H_0 does not matter here (see I). This has the Holstein-Primakoff form plus linear terms in the $k = 0$ mode, and

$$A_k = \varepsilon_k + \frac{3}{2} D_a S (3 \cos^2 \theta - 1) + \frac{3}{2} D_p S \cos(2\psi) \sin^2 \theta \quad (7a)$$

$$B_k = -(\sqrt{6}/4) [\frac{3}{2} D_a S + \frac{1}{2} D_p S \cos(2\psi) (\cos^2 \theta + 1) + i D_p \sin(2\psi) \cos \theta] \quad (7b)$$

and where the expression for ρ_0 , not important here, can also be found in I. In expression (7a), ε_k is given by

$$\varepsilon_k = z - JS \sum_\delta e^{ik \cdot \delta} \quad (8)$$

the summation being extended over the z nearest neighbours (NN), at positions δ , of the probe ion.

The next stage is the full diagonalization of (6), which we will detail here because of the use to be made of it later on. The diagonalization stages are:

$$(i) \quad a_k = \alpha_k + c_k \delta_{k,0} \quad (8a)$$

which eliminates the linear terms in H' ;

$$(ii) \quad \alpha_k = (1/\sqrt{2}) e^{i\Phi_k} (\tilde{\beta}_k + \tilde{\beta}_{-k}) \quad \alpha_{-k} = (1/\sqrt{2}) e^{-i\Phi_k} (\tilde{\beta}_k - \tilde{\beta}_{-k})$$

and cc

$$(8b)$$

which separate out the $+k$ and $-k$ terms; and

$$(iii) \quad \tilde{\beta}_k = l_1 \beta_k + l_2 \beta_k^+ \quad \tilde{\beta}_{-k} = l_1 \beta_{-k} - l_2 \beta_{-k}^+ \quad \text{and cc} \quad (8c)$$

which give rise to a fully diagonalized magnon Hamiltonian, of the form

$$H' = H'_0 + \sum_k [(\beta_k^+ \beta_k + \frac{1}{2}) \tilde{\epsilon}_k - \frac{1}{2} A_k] \quad (9)$$

where

$$\tilde{\epsilon}_k = (A_k^2 - |B_k|^2)^{1/2} \quad (10)$$

are the magnon excitation energies (again, the form of H'_0 does not matter here). The diagonalizing matrices appearing in (8c) are

$$l_{1,2} = ([A_k \pm (A_k^2 - |B_k|^2)^{1/2}]/2(A_k^2 - |B_k|^2)^{1/2})^{1/2} \quad (11a)$$

where the \pm signs in (11a) respectively refer to l_1 and l_2 . The form of c_0 is quite complex (see I), but restricting ourselves here to the case of interest to be considered later on, i.e. SR for $\psi = 0$ (i.e. in plane (c, a)), it has the form

$$c_0 = \sqrt{N} \sqrt{(2S) \sin(2\theta) / \{ [\frac{3}{2}(3 \cos^2 \theta - 1) - (3\sqrt{6}/4) \sin^2 \theta - \Delta [(\sqrt{6}/4)(\cos^2 \theta + 1) - \frac{3}{4} \sin^2 \theta] \}} \quad (11b)$$

where we have introduced the important parameter in this model, $\Delta \equiv -(D_p/D_a)$.

This completes the diagonalization procedure for H . The next stage is the determination of the equilibrium SR angle θ and its temperature dependence, which was fully done in I by minimization of the system free energy, $F = -(1/\beta) \ln Z$, with $\beta \equiv 1/k_B T$ and Z the partition function. Details are given in I. With this background we are able to proceed now to the calculation of the spin correlation functions.

3. Rotation to the magnetization axes frame of spin correlation functions

The situation of the definitions of the Racah or Stevens angular momentum operators in nowadays somewhat intricate and confusing (Rudowicz 1985). Therefore, in order to calculate the spin correlation functions properly, we have to define our operators in a correct and coherent way. We have, as we said before, to calculate the transverse (to the c axis) correlation functions $\langle S_i^x S_m^x \rangle$ and $\langle S_i^y S_m^y \rangle$, as well as the longitudinal one $\langle S_i^z S_m^z \rangle$, i.e. measured relative to the crystal axes. Here $\langle \dots \rangle$ means the thermal average over the energy levels of the already diagonalized Hamiltonian H' , given by equation

(9). Now, rotation by the Eulerian angles, defined in section 2, gives, in the classical limit (infinite S), the following transformation of spin vector coordinates:

$$\begin{aligned} S^x &= S^{x'} \cos \theta \cos \psi + S^{z'} \sin \theta \cos \psi - S^{y'} \sin \psi \\ S^y &= S^{x'} \cos \theta \sin \psi + S^{z'} \sin \theta \sin \psi + S^{y'} \cos \psi \\ S^z &= S^{z'} \cos \theta - S^{x'} \sin \theta. \end{aligned} \quad (12)$$

Here the (x, y, z) indices refer to the crystal axes frame and the primed ones to the rotated magnetization one. Now, for the Rudowicz (1985) and Buckmaster *et al* (1972) transformation of the extended Stevens operators (see section 2) to be coherent with the classical limit, we have to define the first-order Buckmaster (1962) operators in the form

$$\bar{O}_1^0 = S^z \quad O_1^1 = +S^+ \quad O_1^{-1} = +S^- \quad (13)$$

and therefore

$$S^x = \frac{1}{2}(S^+ + S^-) = \bar{O}_1^1 \quad S^y = \frac{1}{2}(S^+ - S^-) = \hat{O}_1^1. \quad (14)$$

The rotation of \bar{O}_1^0 , \bar{O}_1^1 and \hat{O}_1^1 then becomes (Rudowicz 1985, Buckmaster *et al* 1972)

$$[\bar{O}_1^0]_{cf} = -\sin \theta \bar{O}_1^1 + \cos \theta \bar{O}_1^0 \quad (15a)$$

$$[\bar{O}_1^1]_{cf} = \cos \theta \cos \psi \bar{O}_1^1 + \sin \theta \cos \psi \bar{O}_1^0 - \sin \psi \hat{O}_1^1 \quad (15b)$$

$$[\hat{O}_1^1]_{cf} = \cos \theta \sin \psi \bar{O}_1^1 + \sin \theta \sin \psi \bar{O}_1^0 + \cos \psi \hat{O}_1^1 \quad (15c)$$

where the index *cf* refers to the old crystal axes frame and the primed one to the new magnetization rotated one.

The general correlation functions, referred to the crystal axes, are now immediately obtained from (14) and (15). However, we will make the mentioned assumption that the SR takes place on the (a, c) plane of the uniaxial crystal, and make $\psi = 0$ in (15). In such a situation, the spin correlation functions become:

(i) Transverse ($\perp c$)

$$\begin{aligned} C_{lm}^x &\equiv \langle S_l^x S_m^x \rangle = \cos^2 \theta \langle \bar{O}_1^1(l) \bar{O}_1^1(m) \rangle + \sin^2 \theta \langle \bar{O}_1^0(l) \bar{O}_1^0(m) \rangle \\ &\quad + \frac{1}{2} \sin(2\theta) \langle \bar{O}_1^1(l) \bar{O}_1^0(m) + \bar{O}_1^0(l) \bar{O}_1^1(m) \rangle \\ &= \cos^2 \theta \langle S_l^{x'} S_m^{x'} \rangle + \sin^2 \theta \langle S_l^{z'} S_m^{z'} \rangle + \frac{1}{2} \sin(2\theta) \langle S_l^{x'} S_m^{z'} + S_l^{z'} S_m^{x'} \rangle \end{aligned} \quad (16a)$$

$$C_{lm}^y \equiv \langle S_l^y S_m^y \rangle = \langle \hat{O}_1^1(l) \hat{O}_1^1(m) \rangle = \langle S_l^{y'} S_m^{y'} \rangle. \quad (16b)$$

(ii) Longitudinal ($\parallel c$)

$$\begin{aligned} C_{lm}^z &\equiv \langle S_l^z S_m^z \rangle = \sin^2 \theta \langle \bar{O}_1^1(l) \bar{O}_1^1(m) \rangle + \cos^2 \theta \langle \bar{O}_1^0(l) \bar{O}_1^0(m) \rangle \\ &\quad - \frac{1}{2} \sin(2\theta) [\langle \bar{O}_1^1(l) \bar{O}_1^0(m) \rangle + \langle \bar{O}_1^0(l) \bar{O}_1^1(m) \rangle] \\ &= \sin^2 \theta \langle S_l^{x'} S_m^{x'} \rangle + \cos^2 \theta \langle S_l^{z'} S_m^{z'} \rangle - \frac{1}{2} \sin(2\theta) \langle S_l^{x'} S_m^{z'} + S_l^{z'} S_m^{x'} \rangle. \end{aligned} \quad (16c)$$

4. Transverse correlation functions $\langle S_l^x S_m^x \rangle$ and $\langle S_l^y S_m^y \rangle$ and longitudinal $\langle S_l^z S_m^z \rangle$

The averages in equations (16) were calculated, as we have already said, within the spin-wave (sw) approximation. We have, essentially, to calculate diagonal (in x', y', z'

indices) correlation functions $\langle S^i(l)S^i(m) \rangle$, $i = x, y, z$, and the crossed one $\langle S^x(l)S^z(m) \rangle$. Omitting, for simplicity, the primed indices in the spin deviation operators, the correlation functions adopt in the site a_i^\dagger, a_i representation the forms

$$\langle S_i^x S_m^x \rangle = \frac{1}{2} S \langle a_i^\dagger a_m^\dagger + a_i a_m^\dagger + a_i^\dagger a_m + a_i a_m \rangle \quad (17a)$$

$$\langle S_i^y S_m^y \rangle = \frac{1}{2} S \langle -a_i^\dagger a_m^\dagger + a_i a_m^\dagger + a_i^\dagger a_m - a_i a_m \rangle \quad (17b)$$

$$\langle S_i^z S_m^z \rangle = S^2 - \langle a_i^\dagger a_i + a_m^\dagger a_m \rangle + \langle a_i^\dagger a_i a_m^\dagger a_m \rangle \quad (17c)$$

$$\langle S_i^x S_m^z \rangle = \langle S_i^z S_m^x \rangle = \frac{1}{2} \sqrt{(2S)} \langle S(a_i + a_i^\dagger) - a_i a_m^\dagger a_m - a_i^\dagger a_m^\dagger a_m \rangle. \quad (17d)$$

Only the terms of the kind $\langle a_i a_m \rangle$, $\langle a_i^\dagger a_m \rangle$, $\langle a_i a_m^\dagger a_m \rangle$ and $\langle a_i^\dagger a_i a_m^\dagger a_m \rangle$ and complex conjugates (CC) are distance-dependent, giving rise to k -dependent measurable structure factors by SANS techniques. There are also distance-independent terms that, as we shall see, give rise to ferromagnetic long-range contributions to the correlation functions. The averages have to be computed in the representation of wavefunctions β_k^\dagger, β_k , where the overall Hamiltonian H is diagonal (see equation (9)).

The computation of the averages [17a-d] is quite straightforward, and therefore we will only indicate here the final results, leaving for the appendix the details of the main stages of the calculations. They all have the common reciprocal-space Fourier transform

$$F(l-m) = \frac{1}{N} \sum_k \{I_2(k) + [I_1^2(k) + I_3^2(k)] \langle \beta_k^\dagger \beta_k \rangle\} \cos[k \cdot (l-m)] \quad (18)$$

which is paramount in this work. Clearly the first term represents, in a SANS experiment, the scattering due to zero-point quantum fluctuations, and the second one the scattering produced by thermally excited magnons. We should mention that in a series of SANS experiments done on the pseudoternary compounds $(\text{Er}_x\text{Nd}_{1-x})_2\text{Fe}_{14}\text{B}$, only the first contribution has been observed, the magnon effect being negligible (del Moral *et al* 1991).

We will summarize now the form of the correlation functions (17a-d), on the rotated magnetization frame (see appendix):

$$\langle S_i^x S_m^x \rangle = 2Sc_0^2/N + \frac{1}{2} SF(l-m) \quad (19a)$$

$$\langle S_i^y S_m^y \rangle = \frac{1}{2} SF(l-m) \quad (19b)$$

$$\langle S_i^z S_m^z \rangle = \sqrt{2}S^{3/2}c_0/\sqrt{N} - \frac{1}{2}\sqrt{(2S)}(c_0/\sqrt{N})F(l-m) \quad (19c)$$

$$\langle S_i^x S_m^z \rangle = S^2 - 2c_0^2/N + c_0^4/N^2 + (2c_0^2/N - 1)F(0) + (c_0^2/N)F(l-m). \quad (19d)$$

Finally, the measurable, on the crystal frame of axes, spin correlation functions are obtained by combining equations (16) and (19), becoming

$$\begin{aligned} C_{lm}^x &= 2S(c_0^2/N) \cos^2 \theta + [S^2 - 2c_0^2/N + c_0^4/N^2 + (2c_0^2/N - 1)F(0)] \sin^2 \theta \\ &\quad + \sqrt{2}S^{3/2}(c_0/N) \sin(2\theta) + [\frac{1}{2}S \cos^2 \theta + (c_0^2/N) \sin^2 \theta \\ &\quad - \sqrt{2}S^{3/2}(c_0/\sqrt{N}) \sin(2\theta)]F(l-m) \end{aligned} \quad (20a)$$

$$C_{lm}^y = \frac{1}{2} SF(l-m) \quad (20b)$$

$$\begin{aligned}
 C_{lm}^z = & 2S(c_0^2/N) \sin^2 \theta + [S^2 - 2c_0^2/N + c_0^4/N^2 + (2c_0^2/N - 1)F(0)] \cos^2 \theta \\
 & - \sqrt{2}S^{3/2}(c_0/\sqrt{N}) \sin(2\theta) + [\frac{1}{2}S \sin^2 \theta + (c_0^2/N) \cos^2 \theta \\
 & + \sqrt{2}S^{3/2}(c_0/\sqrt{N}) \sin(2\theta)]F(l - m).
 \end{aligned}
 \tag{20c}$$

Finally for a crystal the overall spin correlation function will be

$$C_{lm} \equiv \langle S_l \cdot S_m \rangle = \sum_i C_{lm}^i.$$

However, our main task, in order to give results amenable to comparison with experiments, is the calculation of the reciprocal-space structure factors, transverse, $S^{xz}(\mathbf{k})$ and $S^{yy}(\mathbf{k})$, and longitudinal, $S^{zz}(\mathbf{k})$, and this will be performed in the next section.

5. Small-angle neutron scattering magnetic structure factors

As we have shown above, all the distance-dependent correlation functions have the common function of equation (18), except for proportionality factors. We will now compute $F(l - m)$ passing to the integral approximation and assuming for the magnon exchange energy the harmonic approximation, i.e. $\epsilon_k = Ak^2$, where A is the sw magnon stiffness constant, $A = \frac{1}{2}JSa^2$. This approximation is good for two reasons: We are mainly interested in calculating the structure factors for small-angle neutron scattering experiments, where the momentum transfer is small. Secondly, for the systems alluded to in section 1, the SR regime happens below the SR temperatures T_{SR} , which are low in comparison with the Curie ones, typically $T_{SR} \approx T_C/2$ (Ibarra *et al* 1989a, Marquina 1990).

5.1. Zero-point structure factor

With such assumptions the zero-point quantum fluctuations contribution to $F(l - m)$ will become

$$F_0(l - m) = \frac{v_a}{8\pi^3} \int l_2^2(\mathbf{k}) d^3k = \frac{v_a}{16\pi^3} \int \frac{A_k - (A_k^2 - |B_k|^2)^{1/2}}{(A_k^2 - |B_k|^2)^{1/2}} \cos[\mathbf{k} \cdot (l - m)] d^3k
 \tag{21}$$

where A_k and B_k are given by equations (7), and v_a is the lattice volume per spin. Therefore

$$F_0(l - m) = \frac{v_a}{16\pi^3} \int \left(\frac{\epsilon_k + \frac{3}{2}S\psi_1 - [(\epsilon_k + \frac{3}{2}S\psi_1)^2 - \frac{3}{2}S^2\psi_2^2]^{1/2}}{[(\epsilon_k + \frac{3}{2}S\psi_1)^2 - \frac{3}{2}S^2\psi_2^2]^{1/2}} \right) \cos[\mathbf{k} \cdot (l - m)] d^3k
 \tag{22}$$

where the functions ψ_1 and ψ_2 are defined as

$$\begin{aligned}
 \psi_1(\theta) &= D_a(3 \cos^2 \theta - 1) + D_p \sin^2 \theta \\
 \psi_2(\theta) &= \psi_1(\theta) - 2(D_a + D_p).
 \end{aligned}
 \tag{23}$$

Therefore, the structure factor is given by the expression within the large parentheses. However, this expression is so complicated that it is of little value for comparison with

experiment. However, in the large exchange limit, i.e. $Ak^2 \gg D_a, D_p$, the square roots can be series-expanded and a number of straightforward transformations yield

$$F_0(l-m) = \frac{3v_a}{128\pi^3} \left(\frac{\psi_2}{A}\right)^2 \int \frac{\cos[k \cdot (l-m)]}{(k^2 + \frac{3}{2}S\psi_1/A)^2 - \frac{3}{8}(\psi_2/A)^2} d^3k. \quad (24)$$

Now, if $\psi_2/A \ll 1$ (large exchange limit assumption), we obtain finally

$$F_0(l-m) \approx \frac{3v_a}{128\pi^3} \left(\frac{\psi_2}{A}\right)^2 \int \frac{\cos[k \cdot (l-m)]}{(k^2 + \xi^{-2})^2} d^3k \quad (25)$$

which gives a squared Lorentzian (L^2) structure factor,

$$[\zeta(k)]_0 = (3v_a/2^{11/2}\pi^{3/2})(\psi_2/A)^2/(k^2 + \xi^{-2})^2 \quad (26)$$

with magnetic correlation length

$$\xi = ((2/3S)A/\psi_1)^{1/2} = (2A/3SD_a)^{1/2}[(3\cos^2\theta - 1) - \Delta\sin^2\theta]^{-1/2}. \quad (27)$$

Within the present approximations, this is essentially controlled by the pure classical limit anisotropy free energy (see paper I and section 5.3 below).

5.2. Magnon excitation effects on the structure factor

According to equations (18) and (11a), the magnon excitation contribution to the structure factor is, written in integral approximation,

$$F_T(l-m) = \frac{v_a}{8\pi^3} \int \frac{A_k}{(A_k^2 - B_k^2)^{1/2}} \frac{1}{\exp(\beta\tilde{\epsilon}_k) - 1} \cos[k \cdot (l-m)] d^3k. \quad (28)$$

Again, the structure factor is given by a complicated expression. We will therefore restrict ourselves to the high-temperature limit, i.e. when $\tilde{\epsilon}_k/k_B T \ll 1$ (which constitutes also the most interesting regime, T smaller than but close to T_{SR}), and write down (28) by expanding $\exp(\beta\tilde{\epsilon}_k)$ in the approximate form

$$F_T(l-m) \approx (k_B T) \frac{v_a}{8\pi^3} \int \frac{A_k^2 + \frac{3}{2}S\psi_1(\theta)}{A_k^2 - B_k^2} \cos[k \cdot (l-m)] d^3k. \quad (29)$$

This expression can be transformed by taking into account equations (7a, b), (23) and (27) to

$$F_T(l-m) = \frac{v_a}{8\pi^3} \left(\frac{k_B T}{A}\right) \int \frac{k^2 + \xi^{-2}}{(k^2 + \xi^{-2})^2 - \frac{3}{8}S^2(\psi_2/A)^2} \cos[k \cdot (l-m)] d^3k. \quad (30)$$

In the high exchange limit $(\psi_2/A)^2$ will be small, and therefore we can write (30) in the final form

$$F_T(l-m) = \frac{v_a}{8\pi^3} \left(\frac{k_B T}{A}\right) \int \frac{1}{k^2 + \xi^{-2}} \cos[k \cdot (l-m)] d^3k \quad (31)$$

and therefore the temperature-dependent structure factor becomes

$$[\zeta(k)]_T = (v_a/2\pi^{3/2})(k_B T/A)/(k^2 + \xi^{-2}) \quad (32)$$

i.e. of Lorentzian (L) kind.

5.3. SANS correlation length and domain walls

At $k \neq 0$, long-range ferromagnetic order scattering will not be observed in a SANS experiment and we should look at the origin of the $k > 0$ scattering represented by $[\zeta(k)]_0$ and $[\zeta(k)]_T$. The clue is the obtained correlation length, given by equation (27). For a system described by Hamiltonian (1), the magnetocrystalline anisotropy free energy (per ion), in the classical limit, is precisely (see paper I)

$$\begin{aligned} f_c &= -(S/2)(S/2 - 1)[D_a(3 \cos^2 \theta - 1) + D_p \sin^2 \theta] \\ &= (S/2)(S/2 - 1)[-2D_a + (3D_a - D_p) \sin^2 \theta] \end{aligned} \quad (33)$$

which represents an effective uniaxial anisotropy, of easy c axis if $D_a > D_p/3$, with anisotropy constant

$$K_u = N(S/2)(S/2 - 1)(3D_a - D_p) = N(S^2/4)(3D_a - D_p)$$

for large S , with N being the number of spins per unit volume. For a magnetic domain within the canted magnetization phase, $-N(S/2)(S/2 - 1)\psi_1$ therefore represents the equilibrium anisotropy free energy, and therefore the 180° domain walls separating two domains will have a width (Chikazumi 1964)

$$\delta = \pi(A'/|K_u| \sin^2 \theta(T))^{1/2} \quad (34)$$

where $A' = AS/2N$. If we arbitrarily take the origin of anisotropy energy at $\theta = 0$, it is clear that

$$\delta \approx \pi(A'/NS^2|\psi_1|)^{1/2} \quad (35)$$

and therefore our SANS measured correlation length is, from equation (27), nothing more than

$$\xi \approx 0.2\delta \quad (36)$$

i.e. a length proportional to the domain wall thickness. As we have said, this constitutes the clue to the interpretation of the origin of the SANS scattering: This is the one produced within the domain walls, where ferromagnetic order is destroyed if one thinks of a spiral-like spin configuration. There the spin system is 'disordered' and therefore we find the typical squared Lorentzian structure factor expected for a disordered (e.g. by random direction anisotropy) magnetic system, plus the Lorentzian one also produced in such systems by magnon excitations (Aharony and Pytte 1983, Cullen and del Moral 1992). The key point for the SANS scattering is the uniform ($k = 0$) spin canting, the zero-point quantum fluctuations producing the neutron scattering, but the 'active' volume where such scattering exists being determined by the static ($k = 0$) canting, through the most likely formation of domain walls. In fact, the measured correlation lengths for the $(\text{Er}_x\text{Nd}_{1-x})_2\text{Fe}_{14}\text{B}$ compounds, in the range $\xi \approx 120\text{--}280 \text{ \AA}$ (see section 1), amount roughly to 54 to 125 average lattice constants ($a \approx 11 \text{ \AA}$), which means moderately narrow domain wall widths for such anisotropic materials, but also with high Curie temperatures ($T_C = 600 \text{ K}$) and large spin.

5.4. SANS structure factor for polycrystalline samples

SANS experiments are usually done on polycrystalline samples (del Moral *et al* 1991). Therefore, the magnetic cross section, or the scattered neutron intensity, are proportional to the overall spin magnetic structure factor

$$S_i(\mathbf{k}) = S^{xx}(\mathbf{k}) + S^{yy}(\mathbf{k}) + S^{zz}(\mathbf{k}) \quad (37)$$

which according to equations (20), (26) and (32) are given by

$$S^{xx}(\mathbf{k}) = \frac{v_a}{(2\pi)^{3/2}} f_x \left(\frac{c_0}{\sqrt{N}}, T \right) \delta(\mathbf{k}) + \left(\frac{S}{2} \cos^2 \theta + \frac{c_0^2}{N} \sin^2 \theta - \sqrt{2} S^{3/2} \frac{c_0}{\sqrt{N}} \sin(2\theta) \right) \times \{[\xi(\mathbf{k})]_0 + [\xi(\mathbf{k})]_T\} \quad (38a)$$

$$S^{yy}(\mathbf{k}) = \frac{1}{2} S \{[\xi(\mathbf{k})]_0 + [\xi(\mathbf{k})]_T\} \quad (38b)$$

$$S^{zz}(\mathbf{k}) = \frac{v_a}{(2\pi)^{3/2}} f_z \left(\frac{c_0}{\sqrt{N}}, T \right) \delta(\mathbf{k}) + \left(\frac{S}{2} \sin^2 \theta + \frac{c_0^2}{N} \cos^2 \theta + \sqrt{2} S^{3/2} \frac{c_0}{\sqrt{N}} \sin(2\theta) \right) \times \{[\xi(\mathbf{k})]_0 + [\xi(\mathbf{k})]_T\} \quad (38c)$$

where f_i ($i = x, y, z$) are the distance-independent terms in equations (20a, c). Then, we obtain from (37) and (38a-c)

$$S_i(\mathbf{k}) = \frac{v_a}{(2\pi)^{3/2}} \left[S^2 + 2 \frac{c_0^2}{N} (S - 1) + \frac{c_0^4}{N} + \left(2 \frac{c_0^2}{N} - 1 \right) F(0) \right] \delta(\mathbf{k}) + \left(\frac{3S}{2} + \frac{c_0^2}{N} \right) \{[\xi(\mathbf{k})]_0 + [\xi(\mathbf{k})]_T\}. \quad (39)$$

Therefore the structure factor $S_i(\mathbf{k})$ contains a long-range ferromagnetic component (at $\mathbf{k} = 0$), plus L^2 and L contributions, for finite, although small, wavevectors \mathbf{k} . For the geometry of a SANS experiment, the y component of \mathbf{k} is quenched ($k_y = 0$) and therefore \mathbf{k} is restricted in (39) to the xz plane.

6. Discussion and conclusions

We have shown that below the SR transition temperature T_{SR} , an infinite-range canted ferromagnetic state appears. If the SR is restricted to the (a, c) plane of the crystal axial structure, the associated magnetic cross section is dependent upon the SR angle θ , below T_{SR} . In a SANS experiment, the wavevector \mathbf{k} experimental resolution makes such a contribution invisible, and therefore any possible divergence of the spin correlation length appearing at T_{SR} cannot be detected. In the same form the spin-wave approximation is naturally unable to predict any critical divergence of the long-range spin correlation length, at T_{SR} , which should diverge as $t^{-\nu}$ around T_{SR} ($t = |T - T_{SR}|/T_{SR}$ is the reduced temperature). The SW approximation only shows the existence of the long-range canted ferromagnetic order below T_{SR} . But, besides, the SW approximation predicts the existence of small-angle neutron scattering at finite, but small, wavevectors \mathbf{k} , composed of Lorentzian (L) and squared Lorentzian (L^2) contributions, with a correlation length that is essentially the canted ferromagnetic phase domain wall width. This means that in those competing anisotropy systems, suffering SR transition, the SANS

scattering is produced within the somehow 'disordered' domain walls, and hence the presence of the L^2 component in the structure factor. The L^2 contribution is a direct consequence of the zero-point spin quantum fluctuations, whereas the L contribution is the one from the thermal excitation of magnons.

However, as mentioned in section 1, only the L^2 contribution has been found in the aforementioned $(\text{Er}_x\text{Nd}_{1-x})_2\text{Fe}_{14}\text{B}$ series, probably due to the smallness of the prefactor $(k_B T/A)$ in equation (32), because our SANS experiments were done in such compounds below $T_{\text{SR}} \approx T_C/2$. At small wavevectors $k \neq 0$ our model predicts the existence at T_{SR} of a finite anomaly in the spin correlation length. In fact, from (27), ξ shows a local maximum at T_{SR} , $\xi(T_{\text{SR}}) = (A/3SD_a)^{1/2}$ where $\theta = 0$. This peak has been experimentally observed in SANS experiments of the whole $(\text{Er}_x\text{Nd}_{1-x})_2\text{Fe}_{14}\text{B}$ series of hard intermetallics, as mentioned in section 1, the peak appearing as a very weak (although very well defined) anomaly over a strong background (del Moral *et al* 1991). Notice, from (38), that above T_{SR} ($\theta = 0$) a non-null magnetic scattering is predicted with intensity proportional to $\{3S/2\}[\{\xi(k)\}_0 + \{\xi(k)\}_T]$.

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Appendix

In the following we compute all the site spin deviation operators appearing in the correlation functions considered in this work, in ascending order of the powers of operators involved.

A.1. First-order averages $\langle a_l \rangle$ and CC

We have that

$$\langle a_l^\dagger + a_l \rangle = (1/\sqrt{N})[\langle \alpha_k + c_k \delta_{k,0} \rangle \exp(-i\mathbf{k} \cdot \mathbf{l}) + \langle \alpha_k^\dagger + c_k^\dagger \delta_{k,0} \rangle \exp(-i\mathbf{k} \cdot \mathbf{l})] \\ = 2c_0/\sqrt{N}$$

because $\langle \alpha_k^\dagger \rangle = \langle \alpha_k \rangle = 0$.

A.2. Second-order averages $\langle a_l^\dagger a_m^\dagger \rangle$ and CC

We have

$$\langle a_l^\dagger a_m^\dagger \rangle = \frac{1}{\sqrt{N}} \sum_{k,k'} \langle a_k^\dagger a_{k'}^\dagger \rangle \exp[i(\mathbf{k} \cdot \mathbf{l} + \mathbf{k}' \cdot \mathbf{m})]. \quad (\text{A1})$$

Diagonalizations (i) and (ii) give

$$\langle a_l^\dagger a_m^\dagger \rangle = \frac{1}{\sqrt{N}} \sum_{k,k'} \langle \alpha_k^\dagger \alpha_{k'}^\dagger \rangle + (c_0^\dagger)^2/N = \frac{1}{2N} \sum_{k,k'>0} \exp[-i(\Phi_k + \Phi_{k'})] \\ \times \langle \tilde{\beta}_k^\dagger \tilde{\beta}_{k'}^\dagger + \tilde{\beta}_{-k} \tilde{\beta}_{k'}^\dagger + \tilde{\beta}_k \tilde{\beta}_{-k'} + \tilde{\beta}_{-k} \tilde{\beta}_{-k'} \rangle \exp[i(\mathbf{k} \cdot \mathbf{l} + \mathbf{k}' \cdot \mathbf{m})] + c_0^2/N. \quad (\text{A2})$$

We have to diagonalize now through the (iii) stage; omitting terms whose averages are null, owing to the lack of final diagonal character, we have the following:

(i)

$$\begin{aligned} & \sum_{k,k' > 0} \langle \tilde{\beta}_k^+ \tilde{\beta}_k^+ + \tilde{\beta}_{-k}^+ \tilde{\beta}_{-k}^+ \rangle \exp[-i(\Phi_k + \Phi_{k'})] \exp[i(k \cdot l + k' \cdot m)] \\ &= \sum_{k,k' > 0} (l_1' l_2 \langle \beta_k^+ \beta_{k'} \rangle + l_1 l_2' \langle \beta_{k'} \beta_k^+ \rangle - l_1' l_2 \langle \beta_{-k}^+ \beta_{-k'} \rangle - l_1 l_2' \langle \beta_{-k'} \beta_{-k}^+ \rangle) \\ & \quad \times \exp[i(k \cdot l + k' \cdot m)] \exp[-i(\Phi_k + \Phi_{k'})] = 0 \end{aligned} \quad (A3)$$

because the summation is extended over $k, k' > 0$; l_1, l_2, l_1', l_2' do not depend on the sign of k (see equation (11a)) and the thermal averages are

$$\langle \beta_k^+ \beta_{k'} \rangle = \langle \beta_{-k}^+ \beta_{-k'} \rangle = \langle \beta_k^+ \beta_{k'} \rangle \delta_{kk'} \quad (A4)$$

assuming that the lattice possesses inversion symmetry.

(ii)

$$\begin{aligned} & \sum_{k,k' > 0} \langle \tilde{\beta}_{-k}^+ \tilde{\beta}_k^+ + \tilde{\beta}_k^+ \tilde{\beta}_{-k'}^+ \rangle \exp[-i(\Phi_k + \Phi_{k'})] \exp[i(k \cdot l + k' \cdot m)] \\ &= \sum_{k,k'} (-l_2 l_1' \langle \beta_{-k} \beta_k^+ \rangle + l_1 l_2' \langle \beta_{-k}^+ \beta_{k'} \rangle + l_2 l_1' \langle \beta_{k'} \beta_{-k}^+ \rangle - l_1 l_2' \langle \beta_k^+ \beta_{-k'}^+ \rangle) \\ & \quad \times \exp[-i(\Phi_k + \Phi_{k'})] \exp[i(k \cdot l + k' \cdot m)] = 0 \end{aligned} \quad (A5)$$

for the same reasons as in (i).

Now the term $\langle a_l a_m \rangle$ is the cc of $\langle a_l^+ a_m^+ \rangle$ and therefore is also zero; terms $\langle a_l^+ a_m^+ \rangle$ and $\langle a_l a_m \rangle$ do not contribute to the correlation function, except for a dispersionless contribution, i.e.

$$\langle a_l^+ a_m^+ + a_l a_m \rangle = 2c_0^2/N. \quad (A6)$$

A.3. Second-order averages $\langle a_l^+ a_m \rangle$ and cc

We have

$$\begin{aligned} \langle a_l^+ a_m \rangle &= \frac{1}{N} \sum_{k,k'} \langle a_k^+ a_{k'} \rangle \exp[i(k \cdot l - k' \cdot m)] = \frac{1}{N} \sum_{k,k'} \langle \alpha_k^+ \alpha_{k'} \rangle \\ & \quad \times \exp[i(k \cdot l - k' \cdot m)] + c_0^2/N \end{aligned} \quad (A7)$$

after diagonalization (i). Notice that terms $\langle \alpha_k^+ \rangle = \langle \alpha_{k'} \rangle = 0$. Diagonalization (ii) gives

$$\begin{aligned} \langle a_l^+ a_m \rangle &= \frac{1}{2N} \sum_{k,k' > 0} \langle \tilde{\beta}_k^+ \tilde{\beta}_k^+ + \tilde{\beta}_{-k}^+ \tilde{\beta}_{-k'}^+ + \tilde{\beta}_{-k}^+ \tilde{\beta}_{k'} + \tilde{\beta}_k^+ \tilde{\beta}_{-k'} \rangle \\ & \quad \times \exp[-i(\Phi_k - \Phi_{k'})] \exp[i(k \cdot l - k' \cdot m)]. \end{aligned} \quad (A8)$$

Now, diagonalization (iii) produces interesting consequences, one of which is in the

form of a contribution of zero-point quantum fluctuations to the correlation function. Effectively, extended the summation now to $k, k' > 0$ and $k, k' < 0$, we obtain

$$\begin{aligned} & \frac{1}{2N} \sum_{k,k'} \langle \tilde{\beta}_k^+ \tilde{\beta}_k + \tilde{\beta}_{-k}^+ \tilde{\beta}_{-k'} \rangle \exp[i(\Phi_k - \Phi_{k'})] \exp[i(k \cdot l - k' \cdot m)] \\ &= \frac{1}{2N} \sum_{k,k'} (l_1 l_1' \langle \beta_k^+ \beta_{k'} \rangle + l_2 l_2' \langle \beta_k \beta_{k'}^+ \rangle + l_1 l_1' \langle \beta_{-k}^+ \beta_{-k'} \rangle + l_2 l_2' \langle \beta_{-k} \beta_{-k'}^+ \rangle) \\ & \quad \times \exp[i(\Phi_k - \Phi_{k'})] \exp[i(k \cdot l - k' \cdot m)] \\ &= \frac{1}{2N} \sum_k \left(l_2^2 + \sum_k (l_1^2 + l_2^2) \langle \beta_k^+ \beta_k \rangle \right) \exp[ik \cdot (l - m)] \end{aligned} \quad (\text{A9})$$

considering (A4) and the independence of the $l_i(k)$ matrix on the sign of k .

Therefore, the term $a_l^+ a_m$ and its CC, $a_l a_m^+$, give

$$\langle a_l^+ a_m + a_l a_m^+ \rangle = \frac{1}{N} \sum_k \{ l_2^2(k) + [l_1^2(k) + l_2^2(k)] \langle \beta_k^+ \beta_k \rangle \} \cos[k \cdot (l - m)] + 2c_0^2/N \quad (\text{A10})$$

which is distance-dependent and, as already mentioned, contains a zero-point quantum fluctuations term plus a thermally excited magnon one, and also a distance-independent contribution.

A.4. Third-order averages $\langle a_l a_m^+ a_m \rangle$ and CC

We have

$$\langle a_l a_m^+ a_m \rangle = \frac{1}{N^{3/2}} \sum_{k_1, k_2, k_3} \langle a_{k_1} a_{k_2}^+ a_{k_3} \rangle \exp[i(-k_1 \cdot l + k_2 \cdot m + k_3 \cdot m)]. \quad (\text{A11})$$

Performing, as before, diagonalization (i), disregarding terms $\langle \alpha_k \rangle = \langle \alpha_{k_1} \alpha_{k_2}^+ \alpha_{k_3} \rangle = 0$ (the last one becoming zero after HP diagonalizations (ii) and (iii)), we obtain

$$\begin{aligned} \langle a_l a_m^+ a_m \rangle &= \frac{c_0}{N^{3/2}} \sum_{k_1, k_2} \langle \alpha_{k_1} \alpha_{k_2}^+ \rangle \exp[i(-k_1 \cdot l + k_2 \cdot m)] \\ & \quad + c_0^+ \sum_{k_2, k_3} \langle \alpha_{k_2}^+ \alpha_{k_3} \rangle \exp[i(k_2 - k_3) \cdot m] \\ &= \frac{c_0}{N^{3/2}} \sum_{k_1, k_2} \langle \alpha_{k_2}^+ \alpha_{k_1} \rangle \exp[i(k_2 \cdot m - k_1 \cdot l)] + \delta_{l,m} \\ & \quad + c_0^+ \sum_{k_2, k_3} \langle \alpha_{k_2}^+ \alpha_{k_3} \rangle \exp[i(k_2 - k_3) \cdot m] \end{aligned} \quad (\text{A12})$$

where $\delta_{l,m} = 0$ for $l \neq m$, which is the case. When adding the CC average $\langle a_l^+ a_m a_m^+ \rangle$, it is easy to prove that distance-independent terms cancel out and therefore they need not

be considered any more. The first term in (A12) has already appeared in (A7), and therefore using the previous result,

$$\langle a_l a_m^+ a_m \rangle = \frac{c_0}{2N^{3/2}} \sum_k \{l_2^2(k) + [l_1^2(k) + l_2^2(k)] \langle \beta_k^+ \beta_k \rangle\} \exp[ik \cdot (l - m)]. \quad (\text{A12a})$$

Adding to (A12a) its cc, we finally obtain from (17d)

$$\langle a_l a_m^+ a_m + a_l^+ a_m a_m \rangle = \frac{c_0}{N^{3/2}} \sum_k \left\{ l_2^2(k) + [l_1^2(k) + l_2^2(k)] \langle \beta_k^+ \beta_k \rangle \right\} \cos[k \cdot (l - m)]. \quad (\text{A13})$$

A.5. Fourth-order average $\langle a_l^+ a_l a_m^+ a_m \rangle$

We have that

$$\langle a_l^+ a_l a_m^+ a_m \rangle = \frac{1}{N^2} \sum_{k_1, \dots, k_4} \langle a_{k_1}^+ a_{k_2} a_{k_3}^+ a_{k_4} \rangle \exp[i[(k_1 - k_2) \cdot l + (k_3 - k_4) \cdot m]]. \quad (\text{A14})$$

Performing the diagonalization (i) we immediately obtain the series of equalities

$$\begin{aligned} \langle a_l^+ a_l a_m^+ a_m \rangle &= \frac{c_0^4}{N^2} + \frac{c_0^2}{N^2} \sum_{k_1, k_2} \{2 \langle \alpha_{k_1}^+ \alpha_{k_2} \rangle \exp[i(k_1 - k_2) \cdot l] \\ &\quad + \langle \alpha_{k_1}^+ \alpha_{k_2} \rangle \exp[i(k_1 \cdot l - k_2 \cdot m)] + \langle \alpha_{k_1} \alpha_{k_2}^+ \rangle \exp[-i(k_1 \cdot l - k_2 \cdot m)]\} \\ &\quad + \frac{1}{N^2} \sum_{k_1, \dots, k_4} \langle \alpha_{k_1}^+ \alpha_{k_2} \alpha_{k_3}^+ \alpha_{k_4} \rangle \exp[i[(k_1 \cdot l - k_4 \cdot m) + (k_3 \cdot m - k_2 \cdot l)]] \\ &= \frac{c_0^4}{N^2} + \frac{c_0^2}{N^2} \delta_{l, m} + \frac{c_0^2}{N^2} \sum_{k_1, k_2} \{ \langle \alpha_{k_1}^+ \alpha_{k_2} \rangle \exp[i(k_1 \cdot l - k_2 \cdot m)] + \langle \alpha_{k_2}^+ \alpha_{k_1} \rangle \\ &\quad \times \exp[-i(k_2 \cdot l - k_1 \cdot m)] \} + 2 \frac{c_0^2}{N^2} \sum_{k_1, k_2} \langle \alpha_{k_1}^+ \alpha_{k_2} \rangle \exp[i(k_1 - k_2) \cdot l] \end{aligned} \quad (\text{A15})$$

where we have neglected the fourth-order term in α_k^+ , α_k . The second term is null if $l \neq m$ and the third was already calculated when solving the second-order averages (consult equations (A7) and (A8)). The last one is a particular case of the second one for $l = m$. Therefore, we finally obtain, from (A15),

$$\begin{aligned} \langle a_l^+ a_l a_m^+ a_m \rangle &= \frac{c_0^4}{N^2} + \left(S^2 - 2 \frac{c_0^2}{N} \right) + \frac{1}{N} \left(2 \frac{c_0^2}{N} - 1 \right) \sum_k [l_2^2 + (l_1^2 + l_2^2) \langle \beta_k^+ \beta_k \rangle] \\ &\quad + \frac{c_0^2}{N} \sum_k [l_2^2 + (l_1^2 + l_2^2) \langle \beta_k^+ \beta_k \rangle] \cos[k \cdot (l - m)]. \end{aligned} \quad (\text{A16})$$

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