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Calculations of ferromagnetic surface dynamics using the Green's functions theory and matching procedure

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Abstract. A Heisenberg model is employed to study the spin fluctuation dynamics on a (001) ferromagnetic surface using a new theoretical formalism. The solution of the full magnetic problem arising from the absence of magnetic translation symmetry in one dimension due to the presence of a magnetic surface is presented. The calculations are described using simultaneously a closed form of the spin-wave Green's function and the matching procedure in the random-phase approximation. Analytic expressions for the Green's functions are also derived in a low-temperature spin-wave approximation. The theoretical approach determines the bulk and evanescent spin fluctuation fields in the two dimensional plane normal to the surface. The results are used to calculate the localised modes of magnons associated with the surface. Numerical examples of the modes are given and they are found to exhibit various effects due to the interplay between the bulk and surface modes. It is shown that there may be surface spin-waves that decay in amplitude with distance into the bulk domain. Also the bulk spin fluctuations field as well as the magnons localised at the surface depend on the nature of the bulk-surface coupling exchange. The unstable surface magnetic configurations are illustrated and discussed. The results derived from the dynamic correlation functions between a pair of spin operators at any two sites are employed to evaluate the spin deviation in the ferromagnet due to localised surface modes obtained by the matching procedure as a function of temperature.

PACS. 75.30.Et Exchange and superexchange interactions – 75.50.Dd Nonmetallic ferromagnetic materials – 75.70.Ak Magnetic properties of monolayers and thin films – 76.70.Hb Optically detected magnetic resonance (ODMR)

1 Introduction

Interest in magnetic surfaces from both experimental and theoretical points of view has been motivated by the increasing need to acquire knowledge of their associated magnetic, electronic and mechanical properties for high technology applications. The study of surface spin-waves has proved to be very useful, in particular, for determining a magnetic anisotropy constant [1,2] using Brillouin light scattering [3,4], that provides a tool to probe these magnetic excitations, particularly in ultrathin layers and also in dot-structured permalloy layers [5].

It is well known that localised surface as well as bulk spin-wave modes, may occur in an ordered magnetic solid when the magnetic system does not possess full symmetry translation due to a surface or due to the presence of surface imperfections or other lattice defects such as a surface reconstruction and/or relaxation in the crystal [6–8]. These imperfections may indeed contribute to a number of physical effects such as changes in the thermal properties and the short lifetimes of spin-waves deduced from their observed large linewidths.

There are many reviews of the properties of surface spin-wave in pure ferromagnets including their study by experimental techniques such as light scattering and spin-wave resonance [9,10]. A large number of magnetic excitation investigations of ordered surfaces have been reported in the literature with the aim of describing the magnetic excitations in the case of semi-infinite ferro-and antiferromagnetic cubic systems. The theoretical study of the low-lying magnetic excitations of ordered surfaces has been treated extensively in the harmonic approximation with short-range interactions, using alternatively the Green's functions in the context of the Heisenberg model [11–15], the matching procedure [16] and the transfer matrix approach [17]. Various elegant formulations exist for obtaining the relevant surface Green's functions [18], which can give information on magnetic properties at the crystal surface, spectral densities, influence of surface anisotropy on the magnetisation as well as thermodynamic stability of

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surfaces and the modes of their kinetic growth which are also becoming important.

The matching method which was one of the calculation procedures used with some success to describe vibrational properties of surfaces and resonances [19], was applied to study the surface spin-waves [20]. This approach makes use of a secular equation of spin motion established for the bulk of the semi-infinite magnetic system, which is shown to contain all information concerning the travelling modes, as well as evanescent modes, in the direction perpendicular to the magnetic surface. The eigenvalue matrix is derived from the Heisenberg Hamiltonian, and the energies of the localised states for the surface magnons are obtained using this method, which consists in matching the properties of bulk magnons with evanescent spin-waves on the surface. The existence, the nature and the shape of the solutions are discussed in terms of surface exchange parameters.

Our motivation here is to present some calculations for surface spin-waves using simultaneously the two theoretical approaches based on the close packed Green's functions formalism and the matching procedure. We shall theoretically treat the spin-wave excitations in the simplest case of ferromagnetic media magnetized in the z-direction. The ferromagnetic surface layer is considered to be directly coupled by exchange interactions with the bulk. The localised magnon states occurring at the surface are described. We first obtain explicit Green functions for the surface geometry. These results combined with the matching technique formalism are then employed to calculate the spin-wave states near the surface region.

To illustrate the method that will be employed, we start our study by introducing the total energy including only the first nearest neighbour exchange interactions, magnetic anisotropy and external fields. To do this, the first stage consists of determining, in part, the close packed Green's functions in order to obtain the Bloch equations of spin motions. Secondly, the set of propagating magnetic modes are required, that describe in turn the magnetic propagating bulk spin fluctuations field on the two dimensional (2D) bulk square lattice parallel to the surface structure. This field is independent of the type of surface structure considered, depending only on the nature of the magnetic exchange interactions proposed between its sites. The second stage aims at matching the dynamics of the surface domain to the evanescent bulk spin fluctuations field obtained by means of a Green's function formalism and the matching procedure [21] on the same 2D square lattice as mentioned above. The procedure developed in this paper allows a study of the bulk spin fluctuations field as well as the magnons localised at the surface depending on the nature of the bulk-surface coupling exchange. This generalisation to two dimensions permits the calculation of the magnetic excitation energies that are travelling from the surface to the bulk region.

The outline of this paper is as follows: the theoretical aspects for the bulk magnetic excitations as well as the geometrical model are presented in Section 2. In Section 3, the spin fluctuation dynamics using Green's functions method are presented for the bulk and surface regions, with a view to determining unique solutions for the set of propagating modes on a 2D square lattice parallel to the surface. Then, the Green's functions and the matching procedure technique are simultaneously applied to describe the bulk spin fluctuation dynamics as well as the localised modes of magnons near the surface in Section 4. A simple cubic ferromagnetic structure is assumed, but the results may be generalised to other lattice structures. Section 5 is devoted to determine the dynamic response of the system in order to evaluate static thermodynamic properties. For this purpose, the calculations of the magnetisation deviation due to the surface localised modes obtained from the matching procedure in different layers is described. Numerical applications of the theory and discussion are given in Section 6, whereas the conclusions are summarised in Section 7.

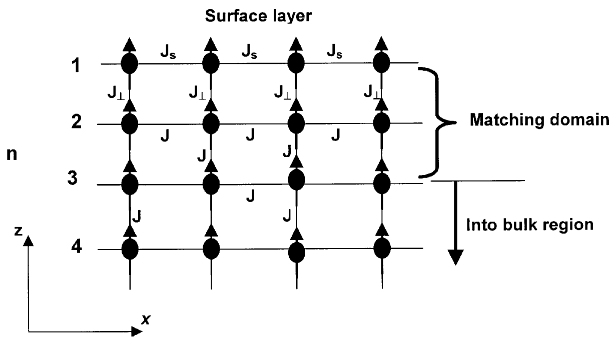

Fig. 1. The geometry considered in the model showing the bulk and surface exchange parameters. The layer number parallel to the surface is indexed by n (along the z-axis). The z axis is normal to the surface boundary, whereas the x and y axis are parallel to the surface layer.

2 Bulk magnetic excitations

The qualitative features of surface spin-waves can be easily illustrated by a (001) face of a simple cubic ferromagnet structure. The crystal is assumed to be infinite in the x and y directions and extends from $z = 0$ to $z = \infty$. The co-ordinates system for the (001) surface is illustrated in Figure 1. The z-axis is chosen to be inwardly normal to the surface plane. Layers parallel to the surface are numbered in ascending order, with the surface as the first layer. The integer n is used to label the layers along the z-axis as indicated in Figure 1. Only the nearest neighbours exchange interactions are considered between the spins in the model assuming that this is sufficient to depict the ground state energy in a Heisenberg Hamiltonian, since these interactions are related to the rapidly decaying electronic wave-function overlap integrals between crystallographic sites. To illustrate this approach, we use the effective spin Heisenberg Hamiltonian including the external and anisotropy magnetic fields to describe the above system. This leads to:

$$H = -\sum_{r \neq r'} J(r, r') S(r) S(r') - \mu H_{a(n)} \sum_{\langle r \rangle} S^Z(r), \qquad (1)$$

$\mu = g\mu_b$ is the gyromagnetic ratio for a magnetic atomic site. $S(r)$ is the local-spin operator located at the lattice site r. $J(r, r')$ denotes the nearest exchange interaction between r and r' sites and is zero except for coupling between nearest-neighbour sites. Also, we allow for the possibility that exchange interactions involving one or more sites in the surface layer may differ from those in the bulk ferromagnet. Beyond the second layer, all exchange interactions have a positive bulk value J. The first nearest neighbours exchange interactions between spins on the surface take the value J_s, whereas $J_\perp$ denotes the interactions between the first and second layers. $H_{a(n)} = H_0 + H^a_{(n)}$ are effective fields experienced by the magnetic atoms in the ferromagnet, due to the externally applied field H_0 and to the effective single-ion anisotropy field $H^a_{(n)}$ acting on the nth layer. They are taken to lie along the easy direction of magnetisation which itself is taken to be the direction of the z-axis, perpendicular to the surface. The values of $H^a_{(n)}$ are considered as $H^a_{(1)}$ on the surface layer and H^a otherwise.

The energy modes of spin-waves E of the retarded Green functions for a Heisenberg ferromagnet have been evaluated using the random phase approximation (RPA). Very briefly, the calculations make use of the general Green's function equation of motion:

$$EG(P:Q) = 1/2\pi\langle[P,Q]\rangle + G([P,H];Q), \tag{2}$$

with H given by (1) and P and Q as the components of the site spin operator.

The resultant series of closed simultaneous equations connecting spin operators on any site in the ferromagnetic crystal is solved using equations (1) and (2), to give the bulk energy spectrum branches for the magnon mode on the ferromagnet and the Green's functions. Assuming isotropic exchange, we find the energy mode to be:

$$E_k = \mu H_a + 2J(r,r')z_{\sigma\kappa}\left\langle S^Z\right\rangle(1-\gamma_k^{\sigma\kappa}), \tag{3}$$

with $\gamma_k^{\sigma\kappa} = \frac{1}{z_{\sigma\kappa}}\sum_{\langle\sigma,\kappa\rangle}\exp[\mathrm{i}k(r_\sigma - r_\kappa)]$. The summation $(\sigma,\ \kappa)$ is over nearest neighbours only, and $z_{\sigma\kappa}$ is the number of nearest neighbours sitting on $[\kappa]$ sites to a magnetic atom on an $[\sigma]$ site. E_k denotes the pole of the Green functions which can be calculated taking into account all the nearest neighbours exchange interactions, the results are:

$$G\left(S^+(r), S^-(r')\right) = \frac{\left\langle S^Z\right\rangle}{\pi N}\sum_k \frac{\exp[\mathrm{i}k(r-r')]}{E-E_k}\cdot \tag{4}$$

N is the number of magnetic unit cells containing one spin each, $\langle S^Z\rangle$ is the thermodynamic expectation values of the z component of a single spin and may be calculated by a molecular field model, which involves calculating the surface magnetisation for a ferromagnetic system using the self consistent molecular field equations [22].

The corresponding Green functions to those in equation (4) but with the spin operator occurring in the reverse order, are obtained using the property $G(Q;P)_E = G(P;Q)_{-E}$. Explicitly, for the reverse order of operators, we obtain also

$$G\left(S^-(r), S^+(r')\right) = -\frac{\left\langle S^Z\right\rangle}{\pi N}\sum_k \frac{\exp[-\mathrm{i}k(r-r')]}{E+E_k}\cdot \tag{5}$$

We can show that the Green function results reduce to those obtained at absolute zero by Keffer for the ferromagnet [23]. Keffer employs the Holstein-Primakoff transformations to diagonalize a spin Hamiltonian, like (1) here, into a boson field of non-interacting magnons at $T \approx 0$ K. At this temperature the z component of spin acquires just the maximum allowable spin value, namely spin quantum numbers S, because the static susceptibilities do not contribute. We can write, in this approximation $\langle S^Z(r)\rangle = S$.

To obtain dispersion relation for the spin-wave modes of the semi-infinite system and furthermore, transverse spin correlation functions of the form $\langle S^+(r,t), S^-(r',t')\rangle$, we evaluate the retarded Green's function $\langle\langle S^+(r,t), S^-(r',t')\rangle\rangle$. In other words, it represents the Fourier components $\langle\langle S^+(r), S^-(r')\rangle\rangle$ at energy E satisfying the usual equation of motion as described by equation (2). Explicitly, it may be expressed as

$$\begin{aligned} E\left\langle\left\langle S^+(r); S^-(r')\right\rangle\right\rangle_E &= 1/2\pi\left\langle\left[S^+(r), S^-(r')\right]\right\rangle \\ &\quad + \left\langle\left\langle\left[S^+(r), H\right]; S^-(r')\right\rangle\right\rangle_E\cdot \end{aligned} \tag{6}$$

Using the property of translational invariance in the $(x,\ y)$ plane, we can introduce the Green's functions and their Fourier transformation into an energy E in the usual way [24] as follows:

$$\begin{aligned} G(r,r';t) &= -\mathrm{i}\left\langle \tau S^+(r,t), S^-(r',0)\right\rangle \\ &= \int G(r,r';E)\exp(-\mathrm{i}Et)\mathrm{d}E. \end{aligned} \tag{7}$$

In a low temperature spin-wave approximation, and considering the normalised energy for the propagating magnetic excitation by putting $E = (\hbar w - g\mu_b H_0)/J$, the equation of motion can be linearized using the RPA procedure. Explicitly, the form taken is given by:

$$\begin{aligned} (E - g\mu H_a)G(r,r';E) &= (S_n/\pi)\delta(r-r') \\ &\quad + \sum_{r''} J(r,r'')S_{n''}G(r,r';E) \\ &\quad - \sum_{r''} J(r,r'';E)S_n G(r'',r';E). \end{aligned} \tag{8}$$

To give the full description of the bulk spin fluctuation dynamics as well as the localised modes on the surface, we need for simplicity, to introduce the following parameters as $\varepsilon_{ij}^{\parallel} = J_s/J; \varepsilon_{ij}^{\perp} = J_\perp/J$. We shall later discuss various models for the values of $\varepsilon_{ij}^{\parallel}$ and $\varepsilon_{ij}^{\perp}$. We can deduce from equation (3), the functions $\gamma_k^{rr'(\parallel)}$ and $\gamma_k^{rr'(\perp)}$ which result from the exponential factors when $z_\parallel$ and $z_\perp$ characterise the number of nearest neighbours in the same layer and the number in an adjacent layer, respectively. These may be expressed in the form:

$$\gamma_k^{rr'(\parallel)} = z_\parallel^{-1} \sum_{(same\ layer)} \exp \mathrm{i} k_\parallel (r - r')$$
$$= 1/2[\cos(k_x a) + \cos(k_y a)],$$
$$\gamma_k^{rr'(\perp)} = z_\perp^{-1} \sum_{(adjacent\ layer)} \exp \mathrm{i} k_\parallel (r - r') = 1. \quad (9)$$

Separating the time dependence of the $S_r^+(t)$ variable given by equation (1) can be done by putting

$$S^+(r,t) = S^+(r)\beta^r \mathrm{e}^{-\mathrm{i}\omega t} \quad (10)$$

where β under β^r is the spatial phase factor of a wave for a propagating mode along the direction perpendicular to the surface. Equation (8) may be recast in the characteristic secular equation form

$$A_0 + A_1\beta + A_2\beta^2 = 0 \quad (11)$$

where the coefficients A_0, A_1 and A_2 (with the property $A_0 = A_2$) may be expressed as:

$$A_0 = A_2 = JS \quad \text{and} \quad A_1 = E + 2JS(\cos(k_x a) + \cos(k_y a)) - JSz - g\mu_b (H_0 + H^a).$$

One can show that the phase factor doublet $(\beta_\xi, \beta_\zeta^{-1})$ both verify the polynomial, owing to the Hermitian nature of the bulk dynamics or time reversal symmetry in magnetic lattices [25]. The energies of the bulk spin fluctuation dynamics field are obtained using equation (11), when β satisfies the propagating condition $|\beta| = 1$ which corresponds effectively to propagating Bloch waves in the bulk that are described by real wave-vectors. The projected bulk band magnon structure onto the surface displays bulk band and forbidden gaps characterised by $|\beta| = 1$ and $|\beta| \neq 1$, respectively. For arbitrary values of β, however, equation (11) does not provide on its own the required unique solution for either of these generic spatial phase factors. To obtain these as a function of E and $k_\parallel a$, one needs also to analyse the spin fluctuation dynamics for the surface domain with the aim to satisfy the evanescent condition $\beta < 1$ which shows that two physically acceptable solutions for β from the roots of equation (11) exist and may be retained.

In order to conclude this section, let us note that the bulk Bloch-waves $|E, k\rangle$, with E and k being respectively the energy and the three-dimensional (3D) wave-vector, are expanded over a set $\{\phi_{\sigma(\kappa),\alpha}\}$ of electronic wave-functions where α designates the site inside the 3D unit-cell. The wave-vector k is defined by its projections $k_\parallel$ and k_z onto the surface and perpendicular to it respectively. The bulk domains are divided into regions, each of them characterised by the number of pairs of Bloch waves $|E, k_\parallel, \pm k_z\rangle$ having same $(E, k_\parallel)$. This then completes the description of the non degenerate evanescent spin-wave modes in the bulk domain, and permits the construction of the evanescent field surrounding the surface region.

3 Surface spin fluctuation dynamics

To develop the matching procedure that will be employed, it is necessary to identify three main domains considering that this technique was given that name because its implementation requires the model to be divided in regions all having the same bidimensional (2D) periodicity along the surface. (i) The bulk regions for a ferromagnet, having 3D periodicity where the bulk magnon dispersion curves are first worked out. (ii) A surface region consisting of an arbitrary number of adsorbate, reconstructed or relaxed layers. (iii) An intermediate region of bulk matter corresponding to the ferromagnet, the thickness of which increases with increasing range of inter-layer interactions, and which is used to match the bulk spin fluctuations field with the boundary conditions imposed by the surface. These different regions are illustrated in Figure 1.

To apply the matching procedure in order to calculate the localised modes of magnons at the surface, we need to know the complete set of evanescent modes in the bulk regions, in the normal direction to the surface plane. These will be used to describe the displacement field in the matching region near the surface as indicated in Figure 1. Owing to the periodic character of the bulk region, its displacement field can be reduced to one vector per layer denoted by $|U_n\rangle$ which represents the spin-wave amplitude for the nth layer. To define this quantity, we need to introduce the amplitude of the precessional spin-waves using time and spatial two-dimensional Fourier transform operators $S^+(r,t)$ given in equation (10). It may be expressed, respectively, as

$$S^+(r,t) = \int_{-\infty}^{+\infty} \mathrm{d}w \exp(\mathrm{i}wt) S^+(r,w), \quad (12)$$

$$S^+(r,w) = (2\pi a)^{-2} \int_{-\infty}^{+\infty} \mathrm{d}k_\parallel \exp(\mathrm{i}k_\parallel \rho) U_{n(r)}(k_\parallel, w), \quad (13)$$

$S^+(r,t)$ denotes the well known spin-lowering operator and may be expressed as $S^+(r,t) = S_X(r,t) + \mathrm{i}S_Y(r,t)$. Now $U_{n(r)}(k_\parallel, w)$ represents the spin-wave amplitude corresponding to n-sites layer located at r, and ρ represents the two-dimensional position vector in x-y plane. We denote $r = (\rho, na)$, where the integer n characterises the atomic position in which that quantum spin value resides at r and a measures the layer spacing.

The pure semi-infinite system has translational symmetry parallel to the surface, allowing us to introduce the Fourier transform from $r_\parallel = (x, y)$ to the projection $k_\parallel = (k_x, k_y)$ which is the two-dimensional propagation vector parallel to the surface, thus

$$G(r, r'; E) = 1/\eta \sum_{k_\parallel} M_{m,l}(k_\parallel, E) \exp[\mathrm{i}k_\parallel (r_\parallel - r'_\parallel)]. \quad (14)$$

η is the number of sites in any one layer and $M_{m,l}$ represents the infinite-dimensional matrix elements describing the equations of motion for each nth layer. Then, after inserting equations (12) and (13) in equation (8) and using equation (14), the above nth layer equation of motion can be rewritten in a matrix form as follows:

$$[M]|U_n\rangle = |0\rangle, \quad (15)$$

where $|U_n\rangle$ is the vector column defined by $|U_n\rangle = [U_1, U_2, U_3, \ldots, U_n, U_{n+1}, \ldots]^T$. Explicitly, equation (15) leads to the set of equations expressed in the following form:

$$\begin{aligned}
EU_1 &= -4\varepsilon_{ij}^{\parallel}\left(\gamma_k^{rr'(\parallel)} - 1\right)U_1 - \varepsilon_{ij}^{\perp}\left(\gamma_k^{rr'(\perp)}U_2 - U_1\right) \\
&\quad + \frac{g\mu_b H_{(1)}^a}{JS}U_1, \\
EU_2 &= -4\left(\gamma_k^{rr'(\parallel)} - 1\right)U_2 - \varepsilon_{ij}^{\perp}\left(\gamma_k^{rr'(\perp)}U_1 - U_2\right) \\
&\quad - \left(\gamma_k^{rr'(\perp)}U_3 - U_2\right) + \frac{g\mu_b H^a}{JS}U_2, \\
EU_n &= -4\left(\gamma_k^{rr'(\parallel)} - 1\right)U_n - \left(\gamma_k^{rr'(\perp)}U_{n+1} - U_n\right) \\
&\quad - \left(\gamma_k^{rr'(\perp)}U_{n-1} - U_n\right) + \frac{g\mu_b H^a}{JS}U_n \quad (\text{for } n \geq 3).
\end{aligned} \quad (16)$$

The complete set of the bulk and evanescent $(n_e + n_b)$ modes outside the surface region $(n \geq 2)$ can be expressed using the translation operator properties [26,27], by the following relations:

$$U_n^b = \sum_{j=1}^{(n_e+n_b)} [\beta'(j)]^{n-3} R_j^+ p'(\beta_j), \quad (17)$$

for a magnetic site in the bulk layers, and as

$$U_n^s = \sum_{i=1}^{(n_e+n_b)} [\beta(i)]^{-(n-3)} R_i^- p(\beta_i), \quad (18)$$

for spin sites located at the surface and matching regions $(n \leq 3)$. In this formalism, R_j^+ and R_i^- depict basis unit vectors spanning the space of the solutions corresponding to the set $\{\beta(i), \beta'(j)\}$ which allows a projection of the evanescent field in the surface domain. The coefficient $p(\beta)$ identifies the relative weighting factors associated with the different spin fluctuation variables. This theoretical representation allows one to treat both the localised modes and the vibrational properties for the model for that purpose by generalising the framework of the matching method from one to two dimensions. Let us mention that since the phonon model [28], as well as the spin-wave model presented here, share a common theoretical approach in the harmonic approximation, it seems reasonable to suppose that the calculations presented here are realistic and predict surface localised spin-waves.

Knowledge of a complete set of evanescent modes in the bulk region requires a description of the ferromagnetic bulk spin fluctuations amplitude field $|U_n\rangle$ in the matching region using the mean-field approximation. The evanescent precessional spin amplitude fields in the bulk region, away from the surface region is described by the phase factor doublet $(\beta_\xi, \beta_\zeta^{-1})$ with respect to ξ and ζ symbols carried out over all travelling 3D ($|\beta_\xi| = 1$ and $|\beta_\zeta^{-1}| = 1$) and exponential-like ($|\beta_\xi|\langle 1$ and $|\beta_\zeta^{-1}|\rangle 1$) Bloch waves. In other terms, we consider that an evanescent magnetic excitation from the surface is characterised by a phase factor satisfying the requirement that $|\beta| < 1$ whereas the propagating mode is described when $|\beta| = 1$. In practice only the evanescent and propagating modes are retained as physically applicable.

Using the above matching procedure in two dimensions it is finally possible to express the equations for the dynamics of the spin fluctuation variable near the surface region, after inserting equations (17, 18) in the homogeneous linear system of equation (16), and then by extracting from equation (15) the sub-matrix corresponding to the set of integers which satisfy the condition $n \leq 3$ corresponding to the labelling surface and matching domains. This procedure allows the surface spin fluctuations variables to be obtained in the form of a square matrix $M^s(l, l')$ acting on a column vector V such as $V = (U_1, R^+, R^-)^T$. This leads to the linear homogeneous system

$$\left\lfloor E^2 I - M^s(\upsilon, \theta, \{\beta\})\right\rfloor |V\rangle = |0\rangle. \quad (19)$$

where I denotes the unit matrix and $\vartheta, \theta = \phi(E, \varepsilon_{ij}^{\parallel(\perp)}, \gamma_k^{rr'(\parallel,\perp)}(k_\parallel,\ \beta_\xi),\ p(\beta_{i(j)}),\ H^a)$, whereas $M^s(\vartheta, \theta, \{\beta\})$ characterises the (3×3) mean dynamical matrix which describes magnons localised on the bulk and surface regions. $\{\beta\}$ is a set of $\xi = 1, 2, \ldots, (n_e + n_b)$ roots of the β-secular equation (11), in the $(E, k_\parallel)$ space. A non trivial solution of equation (19) requires that the determinant of this system vanishes, which defines an algebraic equation in E, whose real and positive solutions $E_S(k_\parallel)$ yield the mean surface magnon branches in the n_b zones, and the surface resonances in the regions where $n_b \neq 0$. Consequently, the localised magnon states can be calculated also when the determinant system given by equation (19) vanishes, so $\det(M^s) = 0$. The non vanishing matrix elements M^s are given in the following form:

$$\begin{aligned}
M^s(1,1) &= E + 4\varepsilon_{ij}^{\parallel}\left(\gamma_k^{rr'(\parallel)} - 1\right) - \varepsilon_{ij}^{\perp} - \frac{g\mu_b H_{(1)}^a}{JS}, \\
M^s(1,2) &= M^s(2,1) = \varepsilon_{ij}^{\perp}\gamma_k^{rr'(\perp)}, \\
M^s(2,2) &= E + 4\gamma_k^{rr'(\parallel)} - 5 - \varepsilon_{ij}^{\perp} - \frac{g\mu_b H^a}{JS}, \\
M^s(2,3) &= \gamma_k^{rr'(\perp)} p(\beta_1), \\
M^s(3,2) &= \gamma_k^{rr'(\perp)}, \\
M^s(3,3) &= \left[E - 6 + 4\gamma_k^{rr'(\parallel)} - \frac{g\mu_b H^a}{JS}\right] p(\beta_1)\beta_1 \\
&\quad + \gamma_k^{rr'(\perp)} p(\beta_1)\beta_1.
\end{aligned}$$

Since the matching method allows both acoustic and optical localised surface spin-wave modes to be obtained, we need in order to complete this study to calculate the correlation functions between a pair of spin operators at any two sites within a model for that purpose and to deduce the magnetisation deviation in each layer n due to these localised modes. Interest in calculating the correlation functions has been motivated by the need to acquire knowledge of the dynamic response of the magnetic system using neutron scattering, light scattering or magnetic resonance. Furthermore, they can be employed to deduce static thermodynamic properties.

4 Correlation functions and layer magnetisation

It is well known that at absolute zero temperature, all of the quantities $\langle S_n^Z \rangle \rightarrow S$, where S denotes the magnitude of the spin of the magnetic ion. At low temperatures, the variation of the expectation value $\langle S^Z \rangle$, due to thermal excitations of spin-waves, is small. Furthermore, $\langle S_n^Z \rangle$ is approximately uniform except near the surface. The magnetisation deviation defined by the quantity $\Delta S_n^Z = S - \langle S_n^Z \rangle$ is also twice as large for the surface spin ($n = 1$), than for the bulk spins ($n \rightarrow \infty$).

The transverse spin correlation function $\langle S^+(r,t)S^-(r',t') \rangle$ can be readily derived from $\langle\langle S^+(r,t)S^-(r',t') \rangle\rangle$ using the fluctuation dissipation theorem. We define a Fourier transformation to $k_\parallel$ and E variables by

$$\left\langle S^+(r,t)S^-(r',t') \right\rangle = \frac{1}{\eta} \sum_{k_\parallel} \exp(\mathrm{i}k_\parallel(r_\parallel - r'_\parallel) \times \int_{-\infty}^{+\infty} \exp(-\mathrm{i}E(t-t')K_{m,l}(k_\parallel, E)\mathrm{d}E. \quad (20)$$

The relation between the Fourier transformed spin correlation function $K_{m,l}(k_\parallel, E)$ and the retarded Green function $M_{m,l}(k_\parallel, E)$, given in equation (14), may be expressed as:

$$K_{m,l}(k_\parallel, E) = \frac{2}{\exp(-E/k_B T) - 1} \mathrm{Im}[M_{m,l}(k_\parallel, E)], \quad (21)$$

where T is the absolute temperature. This analysis enables us to deduce the magnetisation deviation in each layer n which may be expressed as $\Delta S_n^Z = S_n^Z - \langle S_n^Z \rangle$. Solving equation (14) to obtain analytic expressions for the Green's functions and equation (19) in order to deduce the localised modes which will be considered as contributing to determine ΔS_n^Z, we find the equal-time transverse spin correlation functions is

$$\Delta S_n^Z = \left\langle S^+(r,t)S^-(r,t) \right\rangle_{k_\parallel} = -\frac{2}{\eta} \sum_{k_\parallel} \int_{-\infty}^{+\infty} \mathrm{d}E[f(E)+1]\mathrm{Im}\left[M_{m,l}^s(k_\parallel, E)\right], \quad (22)$$

where $f(E) = [\exp(E/k_B T) - 1]^{-1}$. By substituting for $M_{m,l}^s(k_\parallel, E)$ into the equation (22) we can deduce the results for ΔS_n^Z numerically. Let us note that in this procedure and for $T \ll T_c$ (T_c is the Curie temperature) as well as for low enough temperatures, the thermal fluctuations in the magnetisation are relatively small. For this reason, we assume that the spin-wave interactions can be neglected in the above calculations.

5 Numerical results and discussion

Here some numerical examples are presented to demonstrate the essential features of the localised surface spin-waves modes for an insulating ferromagnet with spin value $S = 3/2$ and the influence of the surface-bulk exchange coupling. It is important to note that in the present calculations, we neglect in our numerical results the externally applied field and both bulk and surface anisotropy fields. Their eventual inclusion into the theoretical model poses no difficulty at all from a mathematical point of view and allows a treatment of any ferromagnetic model for that purpose by taking into account the experimental values.

The resolution of equations (11) and (19) leads to a non linear expression in E and $k_\parallel$. Its numerical solution, in the form of a set of points E *versus* $k_\parallel$, gives the bulk spin fluctuation fields for A and B ferromagnetic medias and the dispersion curves of magnetic excitations near the surface, respectively. The only regions where $|\beta| = 1$ corresponds effectively to the propagating modes for bulk region as shown by the shaded area in all figures. Near the surface region, the dispersion curves depict magnons propagating along the direction normal to the surface that are however effectively localised in the sense that their spin fluctuation field is evanescent in the plane normal to the surface. The amplitude of the localised spin-waves in the surface region decays exponentially with increasing penetration into the bulk layers.

To find the surface spin-wave dispersions, we must solve equation (19) by considering $\det(M^s) = 0$. When the solutions exist, they lead to a specific relation between E and $k_\parallel$, $E(k_\parallel)$. The particular dependence of E and $k_\parallel$ depends upon the crystal surface and the values of $\varepsilon_{ij}^\parallel$ and $\varepsilon_{ij}^\perp$. A surface wave branch will become degenerate with the top or bottom of the bulk continuum, when $E_B(k_\parallel^c) = E_S(k_\parallel^c) = 0$, where $k_\parallel^c$ is the critical value of $k_\parallel$.

Let us consider, firstly, the situation of the "free surface" layer corresponding to $J_s = J_\perp = J$ ($\varepsilon_{ij}^\parallel = 1$ and $\varepsilon_{ij}^\perp = 1$). This case characterises the absence of surface perturbations. No surface wave branch exists for the simple cubic (001) surface, with nearest-neighbour exchange interactions. When the exchange is greater at the surface, as illustrated in Figure 2, an optical branch does not necessarily occur. We note that when $1 < \varepsilon_{ij}^\parallel < 1.25$, the solution obtained in the numerical calculations corresponds to a wave whose energy occurs in the bulk continuum. It may be attributed to a specific wave which grows with distance from the surface satisfying the condition $|\beta| > 1$, and is therefore physically not retained. In thin films, solutions in this range are not necessarily forbidden. The requirement for an optical mode is $\varepsilon_{ij}^\parallel \geq 1.25$. We note that an optical mode cannot exist at $k_\parallel = 0$ for a (001) surface, if $\varepsilon_{ij}^\perp = 1$, regardless of the value of $J_\parallel$. For a given value of $\varepsilon_{ij}^\parallel > 1.25$, the optical branch will be truncated at some value of $k_\parallel = k_\parallel^c$ (given by $k_\parallel^{cl} \approx 0.8$, $k_\parallel^{c2} \approx 1.1$, $k_\parallel^{c3} \approx 1.5$) corresponding to $E_B(k_\parallel^c) = E_S(k_\parallel^c)$.

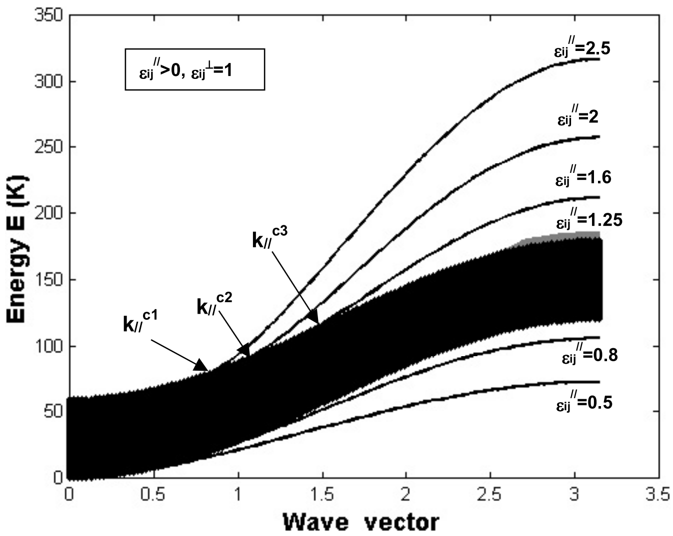

Fig. 2. The figure depicts the bulk and surfaces dispersion modes for $\varepsilon_{ij}^{\perp} = 1$ and for various values of $\varepsilon_{ij}^{\parallel}$. The plot is given against $k_{\parallel}a$ for an in-plane propagating wave-vector $k_{\parallel} = (k_{\parallel}, 0)$. The bulk spin-wave energies are shown shaded, whereas the solid lines represent the surface modes. The energies are given in kelvin units.

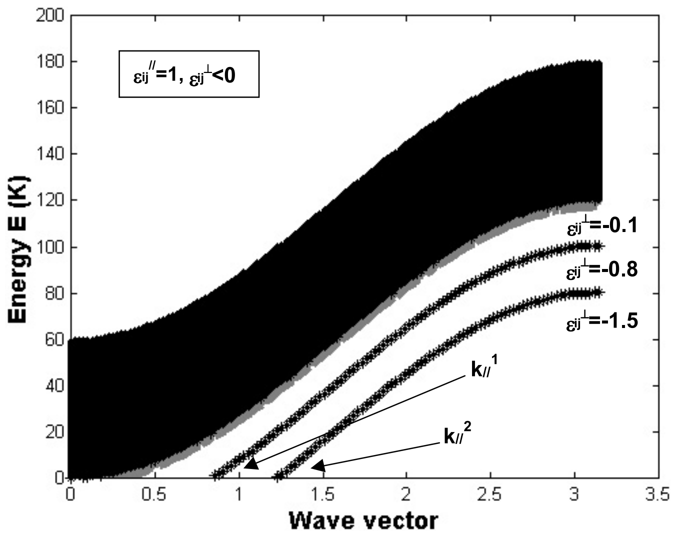

Fig. 3. The figure depicts the numerically calculated dispersion relations considering $\varepsilon_{ij}^{\parallel} = 1$ and for various values of $\varepsilon_{ij}^{\perp}$. The plot is given against $k_{\parallel}a$ for an in-plane propagating wave-vector $k_{\parallel} = (k_{\parallel}, 0)$. The bulk spin-wave energies are shown shaded, whereas the solid lines represent the surface modes. The energies are given in kelvin units.

The investigations of the behaviour of the ferromagnetic system when considering the antiferromagnetic surface exchange are illustrated in Figures 3 and 4 showing the unstable magnetic configuration. In this representation, two simplified models have been used. The first is based on the case when the surface exchange constant $J_{\perp}$ is negative and $J_s = J$. The second example of antiferromagnetic rearrangement occurs when J_s is negative and $J_{\perp} = J$.

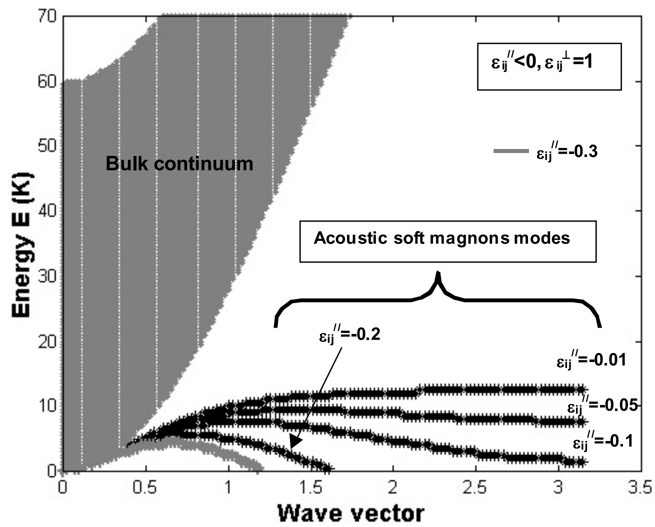

Fig. 4. The energy of surface spin-waves plotted *vs.* $|k_{\parallel}a|$ for an in-plane propagating wave-vector $k_{\parallel} = (k_{\parallel}, 0)$ for various values $\varepsilon_{ij}^{\parallel}$ considering $\varepsilon_{ij}^{\perp} = 1$. The acoustic soft magnons modes are illustrated characterising the unstable arrangements of surface spin fluctuations.

Considering the case in which the constant J_s takes the same value as the positive bulk exchange interaction $J(\varepsilon_{ij}^{\parallel} = 1)$ and $J_{\perp}$ is assumed to be negative ($\varepsilon_{ij}^{\perp} < 0$). The numerical results are presented in Figure 3. In this representation, we have $E_S(k_{\parallel}) < E_B(k_{\parallel})$. Solutions with this property are acoustic surface spin-waves. As $\varepsilon_{ij}^{\perp} \to 0$, $E_S \to 0$ as $k_{\parallel} \to 0$. For $\varepsilon_{ij}^{\perp} = -0.1$, the acoustic spin-wave branch lies at the boundary of the bulk continuum. For $\varepsilon_{ij}^{\perp} > -0.1$, no surface branch occurs. The qualitative features for $\varepsilon_{\parallel}^{\perp} < 0$ can be seen from the cases where $\varepsilon_{ij}^{\perp} < -0.1$. We then find a surface branch in some regions ($k_{\parallel} < k_{\parallel}^1$ and $k_{\parallel} < k_{\parallel}^2$) with the values of E_s being negative (precession in opposite sense). As $k_{\parallel}$ increases, the surface wave becomes more localised at the surface. At the zone edge ($k_x = k_y = \pi/a$), only the surface layer has a non negligible amplitude. Figure 5a illustrates the surface wave eigenvectors for $\varepsilon_{ij}^{\parallel} = 1$ and $\varepsilon_{ij}^{\perp} < 0$ corresponding to an antiferromagnetically coupled surface layer. Only the surface layer has a non negligible amplitude. The phase of the spin fluctuation amplitude components U_n is constant, whereas the magnitude decreases with increasing penetration into the bulk. Let us note that for the limit $k_{\parallel} \to 0$ corresponds to the quantity $E_S \to 0$. If $E_S = 0$ for any $k_{\parallel} \neq 0$, then the assumed ferromagnetic spin arrangement is not the ground state of the system. The unstable mode at $k_{\parallel} = 0$ has all surface spins in phase. In such case, the new ground state must be used to obtain meaningful solutions. It may be also described by a ferromagnetic surface layer configuration, but the spins acting on surface must be aligned antiparallel to the second and subsequent layers.

In the opposite case, the dispersion relations corresponding to $\varepsilon_{ij}^{\parallel} < 0$ and $\varepsilon_{ij}^{\parallel} = 1$ are illustrated in

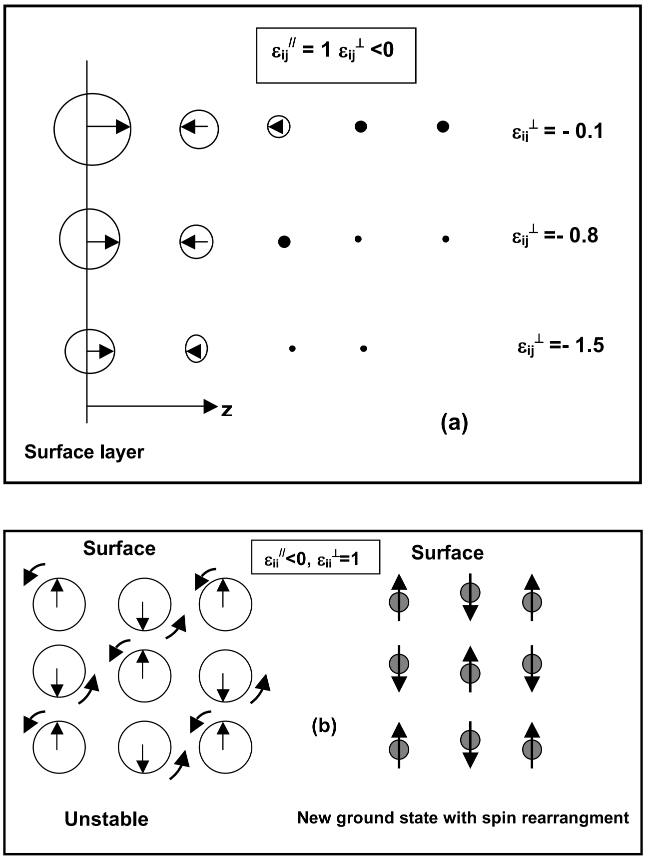

Fig. 5. (a) Surface wave eigenvectors for $\varepsilon_{ij}^{\parallel} = 1$ and $\varepsilon_{ij}^{\perp} < 0$ corresponding to an antiferromagnetically coupled surface layer ($\varepsilon_{ij}^{\perp} < 0$). The ground state has a ferromagnetic surface layer with spins aligned opposite to the remaining ferromagnetic layers. (b) Surface waves for antiferromagnetic surface exchange. The description of unstable mode for $\varepsilon_{ij}^{\parallel} < 0$ on surface layer is illustrated and the new configuration of the surface layer described by the new ground state is given.

Figure 4. The solutions obtained show the presence of acoustic soft modes of magnons with negative slope which occur for $\varepsilon_{ij}^{\parallel} < -0.05$. This characterises the unstable arrangement of spin in surface layer. When $\varepsilon_{ij}^{\parallel} = -0.1$, the corresponding localised spin-wave energy E_s vanishes at the edge of the two-dimensional zone for which $k_x = k_y = \pi/a$. For these wave-vectors, the spins on a given layer are 180° out of phase with their nearest neighbours on the same layer. Since the energy of this mode is zero, the radius of the precessional circle may grow arbitrarily large with no expenditure of energy. Therefore the system is unstable. The surface layer configuration is given in Figure 5b. For $\varepsilon_{ij}^{\parallel} < -0.1$, the assumed ferromagnetic ground state does not correspond to that of the lowest energy state. These features allows another ground state to be defined, depicted by the surface spins with antiferromagnetic configuration as shown in Figure 5b.

The primary objective of introducing the surface antiferromagnetic interactions is to give the complete solutions from the theoretical aspects developed in this paper, in order to discuss the existence, the nature and the shape of these solutions. Let us note that the same forms of the dispersion curves, obtained within the theoretical formalism used here, have been obtained in previous work [12]. If we consider that the exchange interactions in and near the surface are antiferromagnetic in sign, then the possibility of the magnetic analogue of surface reconstructions arises. The spins in the surface region may order in an antiferromagnetic manner, even though the bulk is ferromagnetic. The latter situation occurs in many well known magnetic crystals, and the former may prove of interest for an overlayer on a magnetic substrate, or if appreciable expansion occurs near the surface as previously described [29]. Furthermore, it seems to be important to discuss the properties of a model which exhibits this behaviour from a theoretical point of view. The properties of the model may be examined and can constitute a means to understand some aspects of observations of spin polarised photoemission from the ferromagnetic surfaces.

Solving numerically equation (22) to obtain the poles of the Green functions and equation (19) to deduce the surface localised modes, we present in Figures 6a and b, the salient numerical results of the magnetisation deviations ($\Delta S_n^Z/S$) due to the acoustic and optical surface waves dispersion with the same parameters as in Figure 2 ($\varepsilon_{ij}^{\parallel} = 0.8$ and $\varepsilon_{ij}^{\parallel} = 1.6$), respectively. We emphasise that the results presented here do not include the contribution of the bulk modes, because we have summed only over the poles corresponding to evanescent spin-wave modes for which the spin fluctuation field is described by $|\beta| < 1$. On the other hand, the case where only the surface layer has a surface perturbation ($\varepsilon_{ij}^{\parallel} > 0, \varepsilon_{ij}^{\parallel} = 1$) is retained in these calculations. For some values of temperature, we observe a decreasing magnetisation deviation with increasing layer number n. The evolution of $\Delta S_n^Z/S$ as function of temperature depict the presence of one peak located at $n = 1$ associated with the surface localised modes – one acoustic (Fig. 6a) and one given by the optical mode (Fig. 6b).

The spatial variation in magnetisation deviation is shown to produce an effect similar to a weakening of the exchange constants at the surface. Thus, even the "free surface" ($J_\parallel = J_\perp = J$) has surface perturbations in the exchange interactions. As the temperature increases, the effect of the surface on the spatial variation of the magnetisation becomes more important. The number of layers, whose magnetisation is substantially lower than that of the bulk increases with increasing temperature. These results confirm those obtained by Mills and Maradudin in previous work [11].

In Figures 7 and 8, we present the solutions obtained in the case of the presence of surface perturbations. A variety of possible acoustic and optical surface branches are presented for various values of $\varepsilon_{ij}^{\parallel} > 0$ and $\varepsilon_{ij}^{\perp} > 0$. In Figure 7, the surface spin-wave dispersion curves show that more than one surface branch can occur for given values of $\varepsilon_{ij}^{\parallel}$ and $\varepsilon_{ij}^{\perp}$. Two truncated optical branches occur corresponding to $k_\parallel^{c1} \approx 1.7$ and $k_\parallel^{c2} \approx 2.3$ which gives

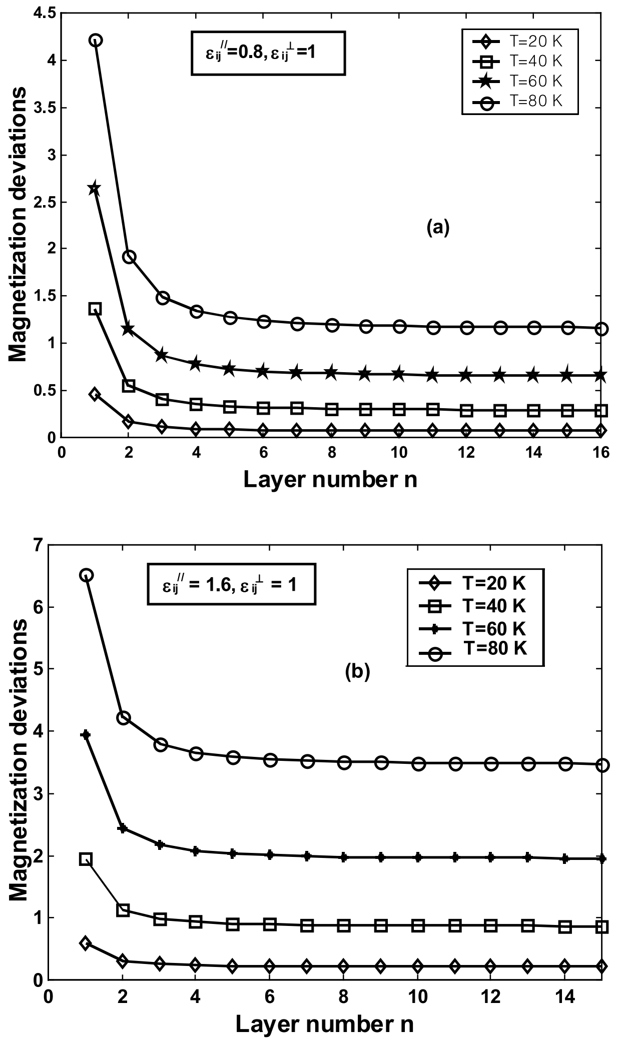

Fig. 6. (a)The magnetisation deviations $[(S - \langle S_n^Z \rangle)/S]$ (with arbitrary units) in layer n for various values of temperatures T. (a) The figure depicts the intensity of the localised spin-waves assuming the acoustic surface localised modes corresponding to $\varepsilon_{ij}^{\|} = 0.8$ and $\varepsilon_{ij}^{\perp} = 1$ (as shown in Fig. 2). (b) The figure depicts the intensity of the localised spin-waves assuming the optical surface localised modes corresponding to $\varepsilon_{ij}^{\|} = 1.6$ and $\varepsilon_{ij}^{\perp} = 1$ (see Fig. 2).

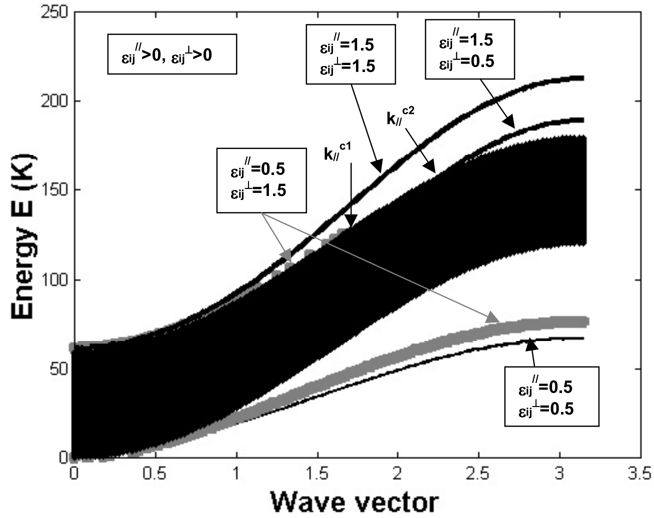

Fig. 7. Bulk and surface dispersion modes for various values of $\varepsilon_{ij}^{\|}$ and $\varepsilon_{ij}^{\perp}$. The plot is given against $k_{\|}a$ for an in-plane propagating wave-vector $k_{\|} = (k_{\|}, 0)$. The bulk spin-wave energies are shown shaded, whereas the solid lines represent the surface modes. More than one surface branch can occur for $\varepsilon_{ij}^{\|} = 0.5$ and $\varepsilon_{ij}^{\perp} = 1.5$.

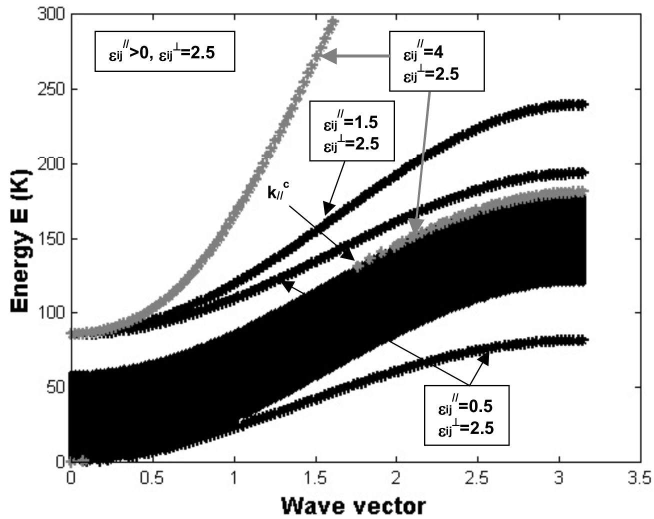

Fig. 8. Bulk and surface dispersion modes for various values of $\varepsilon_{ij}^{\|}$ and $\varepsilon_{ij}^{\perp}$. The plot is given against $k_{\|}a$ for an in-plane propagating wave-vector $k_{\|} = (k_{\|}, 0)$. The bulk spin-wave energies are shown shaded, whereas the solid lines represent the surface modes. For $\varepsilon_{ij}^{\|} = 0.5$ and $\varepsilon_{ij}^{\perp} = 2.5$ a complete optical and a complete acoustic branch occur.

$E_s(k_{\|}^{c1}) \approx 128$ K and $E_s(k_{\|}^{c2}) \approx 158$ K, respectively. Figure 8 illustrates the localised surface magnon modes for a large value of $\varepsilon_{ij}^{\perp} = 2.5$. For $\varepsilon_{ij}^{\|} = 0.5$ and $\varepsilon_{ij}^{\perp} = 2.5$, both complete optical and acoustic branches occur. All the complete optical branches become the origin of the first Brillouin magnetic cell with $E_s(k_{\|} \to 0) \approx 85$ K.

6 Conclusions

The main objective of this work is to demonstrate that it is possible to use simultaneously the matching and Green's function formalism in order to calculate the surface spin-waves modes and the localised modes near the surface domain. In the first part we calculated analytically the spin-spin Green functions in a ferromagnetic structure. This gave results for the spin fluctuations field outside the surface domain. Then, by matching the evanescent bulk spin fluctuation dynamics with the surface modes, we give the spectral intensity of the surface spin-waves, as well as allowing us to recover the well-known surface spin-wave dispersion relations. The surface mode spectral intensity was studied as a function of depth from the surface and in-plane wave-vector for different values of the ratio of surface to bulk exchange $\varepsilon_{ij}^{\parallel}$ and $\varepsilon_{ij}^{\perp}$. It was shown that it is possible for the surface to support both the acoustic and optical modes. These modes depend on the nature and the values of bulk-surface exchange coupling.

In addition, we have expressed the equal-time transverse spin-spin Green functions in the form $\langle\langle S^+(r), S^-(r)\rangle\rangle$ for low temperature $T \ll T_c$. Then, the Green functions and matching method have been simultaneously used to describe the effects of surface spin-waves in a semi-ferromagnet with the aim to evaluate the magnetisation factor ΔS_n^Z due to the surface localised modes as a function of the temperature and layer index n. There are a number of further calculations which may be made using the formalism developed here for the spin-spin Green functions. In particular, the dynamic response of the system due to light scattering or neutron scattering may readily be investigated.

The general description of these spin-wave behaviours has been illustrated by considering the presence or not of the surface perturbations. The analytical approach described for the present case of ferromagnetic insulators can readily be generalized to other surface problems concerning in particular the phonons, itinerant spins or electrons. It can also give in a direct manner the spin fluctuation field on the surface with the help of finite matrices. A straightforward generalization of the present calculations would be to other crystal structures such as fcc and bcc with applications to materials such the EuO and EuS compounds using Brillouin light scattering. These particular materials are relatively strong light scatterers and are relatively opaque, making them suitable for surface studies.

The frequencies of the localised modes described in this work may provide information concerning the local magnetic anisotropy and exchange interactions in the neighbourhood of such a surface, and will contribute to understanding more fully the role played in surface phenomena such as surface instability, the growth of magnetic substrates, and surface optical properties. We emphasize that the present model is simple insofar that it considers only magnetic exchange interactions between the ordered spins, yielding hence the exchange dominated surface localised spin-waves. It is quite possible that other kinds of magnetic interactions also play a role in the behaviour and frequencies of the magnons localised at the surface considering the reconstruction and/or relaxed layers, in which case these interactions should be considered using the equations of motion of the spin fluctuation field.

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