Surface spin dynamics of antiferromagnetically coupled frustrated triangular films

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Results are presented for spin-wave dispersions in geometrically frustrated stacked triangular antiferromagnets with a thin film or semi-infinite geometry having either zero, easy-plane, or easy-axis anisotropy. Surface effects on the equilibrium spin configurations and excitation spectrum are investigated for the case of antiferromagnetically coupled films, serving to extend previous results on ferromagnetically coupled layers [E. Meloche et al., Phys. Rev. B. 74, 094424 (2006)]. An operator equation of motion formalism is applied to systems which are quasi-one- and quasi-two-dimensional in character. In contrast to the case of ferromagnetically coupled films, the results of this work show surface modes that are well separated in frequency from bulk excitations. Magnetic excitations in thin films with an even or an odd number of layers show qualitatively different behavior. These results are relevant for a wide variety of stacked triangular antiferromagnetic materials.

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I. INTRODUCTION

The significant amount of research effort devoted to the discovery and understanding of new aspects of geometrically frustrated magnetic systems in recent decades has been almost exclusively focused on the study of bulk properties.1 Among such systems, a large number of materials have been identified which are realizations of the prototype frustrated triangular antiferromagnet (AF). In addition to the AF near-neighbor in-plane coupling, the vast majority of these systems are also characterized by AF coupling between stacked triangular layers, such as in most of the ABX3 compounds.2 Theoretical and experimental studies of magnetic excitations in these model materials have revealed important information on the nature of the fundamental interactions and exposed new types of spin-wave modes in the bulk systems.3–6 Interest in surface effects on frustrated magnetic systems has been enhanced recently with the fabrication of new material classes. Studies of hexagonal magnetoelectric and multiferroic rare-earth magnetite (e.g., RMnO3) thin films are being fueled by potential technological applications in the field of spintronics.7 Ultrathin films of Mn on fcc (111) Fe-Ni substrates also offer the possibility to study AF triangular layers.8 The understanding of thin-film, surface, and interface spin dynamics of such systems is of interest for the engineering of high data transfer rate devices.9

In the present work, results of solving the operator equation of motion to examine surface spin excitations in a model of AF-coupled frustrated triangular layers which have either easy-plane or easy-axis anisotropy are shown. This study represents an extension of our previous work where the technique was developed and applied to the case ferromagnetically (F) coupled triangular layers having easy-plane anisotropy (see Ref. 11, hereafter referred to as Paper I). A summary of general theoretical and experimental techniques that have been employed to study thin-film and surface spin waves in unfrustrated systems is also provided in Ref. 10. The formulation of a linear spin-wave theory is based on a model Hamiltonian which includes isotropic and anisotropic exchange as well as single-ion anisotropy and allows for layer-dependent coefficients of these interactions. Depending on the specific system of interest (e.g., semi-infinite and thin films with or without adjacent layers of a different material), surface parameters may have values quite different from the bulk. A significant difference found for the present case of AF-coupled layers is that localized modes can be well separated in frequency even with bulk parameter values used at the surface layers, in contrast with F-coupled films. Also, as might be expected, equilibrium spin configurations and excitations are dependent on whether the films are composed of an even or odd number of layers (again in contrast with F-coupled films). Due to physical geometrical considerations, nearly all material realizations of stacked triangular layers can be classified as having either quasi-one-dimensional (quasi-1D) or quasi-two-dimensional (quasi-2D) magnetic character.2 In quasi-1D compounds, the exchange interactions along the crystal c axis are much greater than the weaker AF coupling between nearest-neighbor pairs in the basal plane. In contrast, in quasi-2D compounds, the dominant AF interactions are within the basal plane and the spins along the c axis are weakly coupled. Illustrative numerical results of the spin-wave calculations for these cases are discussed in the following sections with the intent to reveal key aspects of thin-film and surface effects on spin excitations in these systems.

These results are generally applicable to a wide variety of AF-coupled layered triangular materials. Among the ABX3 compounds, CsMnBr3 and CsVCl3 are notable examples of quasi-1D materials with planar anisotropy.3 Bulk excitations have been well studied in prototypical axial quasi-1D hexagonal materials CsNiCl3 and CsCoX3.4 Examples of quasi-2D compounds with well established bulk magnetic properties include axial CuFeO2 (Ref. 5) and planar VX2 and LiCrS2.6 Most of the RMnO3 materials exhibit planar anisotropy although characterization of thin films in this regard is ongoing.7 In ultrathin films, surface effects dominate and spin reorientation transitions can occur.8 A number of possible experimental techniques to examine long wavelength surface modes are summarized in our previous work.11
Progress using inelastic neutron scattering techniques to probe surface excitation in multilayer systems has been reported.\textsuperscript{12}

The systems considered here may be represented using the following spin Hamiltonian:

\begin{equation}
\mathcal{H} = \sum_{i,j} J_{ij} (S_i \cdot S_j + \sigma S_i^z S_j^z) + \sum_{i,j'} J'_{ij} (S_i \cdot S_{j'} + \sigma' S_i^z S_{j'}^z) + \sum_i D_i (S_i^z)^2,
\end{equation}

where \( J_{ij} > 0 \) represents the intralayer nearest-neighbor AF exchange coupling and \( J'_{ij} > 0 \) represents the interlayer AF exchange coupling. The effects of anisotropic exchange coupling are included through the parameters \( \sigma \) and \( \sigma' \), and \( D_i \) represents the strength of the single-ion anisotropy at a magnetic site labeled \( i \). The Hamiltonian in Eq. (1) can be used to describe systems characterized with easy-plane \((D_i > 0)\) and easy-axis \((D_i < 0)\) single-ion anisotropy. Easy-plane and easy-axis systems have different equilibrium spin configurations and spin dynamics and therefore the cases are treated separately. We also allow the intralayer exchange interaction and single-ion anisotropy parameters for spins located at the surface to differ from their respective bulk values. This asymmetry in the surface parameters can be used to investigate the spin-wave properties of a frustrated magnetic thin film placed between different nonmagnetic layers.

In Sec. II, bulk and surface spin-wave modes are calculated for the case of easy-plane anisotropy. In Sec. III, results are deduced for the spin-wave energies and spectral intensities for the case of easy-axis anisotropy. A discussion of these results and their relevance to specific compounds is presented in Sec. IV.

II. EASY-PLANE ANISOTROPY

A. Easy-plane bulk spin-wave excitations

Although the Green’s function formalism used in this work has been developed previously to calculate spin waves in numerous unfrustrated thin-film systems,\textsuperscript{10,13,14} the application to frustrated cases requires some care and a detailed outline is provided here. Extending the general method described in Paper I to calculate the bulk spin-wave dispersion relation, the system is divided into three pairs of sublattices on two adjacent layers, which are labeled \( A_1, B_1, C_1, A_2, B_2, \) and \( C_2, \) and the specific sites on each sublattice are labeled with indices \( l_1, m_1, n_1, l_2, m_2, \) and \( n_2, \) respectively. Within any particular layer, the 120° structure is stabilized as a result of the pairwise antiferromagnetic coupling between nearest-neighbor spins. Also, in the classical ground state configuration, the spins on adjacent layers (along the crystallographic \( c \) axis) order antiparallel to one another. We first transform to a local coordinate system such that the \( z \) axis for each sublattice is in the direction of the average spin alignment. The transformations to the local coordinate systems for the various sites may be written as

\[ (S_i^x, S_i^y, S_i^z) \rightarrow \left[ (1)^{\mu+1}(S_i^x \cos \theta_i - S_i^z \sin \theta_i), (1)^{\mu+1}ight. \]

\[ \times (S_i^y \sin \theta_i + S_i^z \cos \theta_i), -S_i^y^\prime \right], \]

with \( i = l_{\mu}, m_{\mu}, n_{\mu}, \theta_{\mu} = 0, \theta_{\mu} = 2\pi/3, \) and \( \theta_{\mu} = -2\pi/3, \) with \( \mu = 1, 2. \) The equations of motion for the local spin raising and lowering operators at each lattice site are formed using

\[ \frac{idS_i^z}{dt} = [S_i^z, \mathcal{H}], \]

where \( \mathcal{H} \) is the transformed Hamiltonian. The exchange terms on the right-hand side of Eq. (3) involving the product of spin operators at different sites are decoupled using the random-phase approximation. The terms involving the single-ion anisotropy involve the product of spin operators at the same site and these are decoupled using \( (S_i^x S_i^y + S_i^y S_i^x) \rightarrow 2S_p S_i^z, \) where the decoupling parameter is \( p = 1 - (2S)^{-1}. \)

The set of linearized coupled equations is then transformed to a wave vector representation. Assuming the usual time dependence \( \exp(-iEt) \), the resulting set of 12 equations may be expressed as \( \mathbf{M} \mathbf{b} = 0, \) where

\[ \mathbf{M} = \begin{pmatrix} \mathbf{A} & \mathbf{\tau} \\ \mathbf{\tau} & \mathbf{A} \end{pmatrix} \text{ and } \mathbf{b} = \begin{bmatrix} b_1 \\ b_2 \end{bmatrix}. \]

The elements of the 6×6 block-circulant matrices \( \mathbf{A} \) and \( \mathbf{\tau} \) are written as

\[ \mathbf{A} = \begin{pmatrix} \bar{A} & \bar{B} & \bar{B}^* \\ \bar{B}^* & \bar{A} & \bar{B} \\ \bar{B} & \bar{B}^* & \bar{A} \end{pmatrix}, \quad \mathbf{\tau} = \begin{pmatrix} \tau(k_x) & 0 & 0 \\ 0 & \tau(k_x) & 0 \\ 0 & 0 & \tau(k_x) \end{pmatrix}. \]

The matrix elements of \( \mathbf{A} \) and \( \mathbf{\tau} \) depend on the energy and system parameters and are defined as

\[ \bar{A} = \begin{pmatrix} E + \Omega & \alpha \\ -\alpha & E - \Omega \end{pmatrix}, \]

\[ \bar{B} = \begin{pmatrix} \beta & \gamma \\ -\gamma & -\beta \end{pmatrix}, \quad \tau(k_x) = SJ'(k_x)/2 \begin{pmatrix} -\alpha' & 2 + \alpha' \\ -2 - \alpha' & \alpha' \end{pmatrix}. \]

where

\[ \Omega = -SJ(0) - SD' - SJ'(0), \]

\[ \alpha = SD', \]

\[ \beta = -S(1/2 + \sigma)J(k)'/2, \]

\[ \gamma = S(3/2 + \sigma)J(k)'/2, \]

\[ D' = [1 - (2S)^{-1}]D. \]

The exchange integrals \( J(k) \) and \( J'(k_x) \) are defined as in Paper I in terms of the lattice constants \( a \) and \( c. \) The elements of the column vector \( \mathbf{b} \) are written as

\[ 214402-2 \]
\[ b_\mu = [S^{\pm}_{A_\mu}(k), S^{\pm}_{B_\mu}(k), S^{\pm}_{C_\mu}(k), S^{\pm}_{D_\mu}(k), S^{\pm}_{E_\mu}(k), S^{\pm}_{F_\mu}(k), S^{\pm}_{G_\mu}(k), S^{\pm}_{H_\mu}(k)]^T, \]

for \( \mu = 1, 2 \). The terms \( S^{\pm}_{A_\mu}(k) \) correspond to the Fourier amplitudes of the spin operators \( S^{\pm}_{B_\mu}(k) \), for \( \mu = 1, 2 \), along with similar definitions for \( S^{\pm}_{C_\mu}(k) \) and \( S^{\pm}_{D_\mu}(k) \). The bulk spin-wave modes correspond to the solutions of \( \det \mathbf{M} = 0 \). Analytical results may be obtained by first block diagonalizing \( \mathbf{M} \) using the transformation \( \mathbf{W} = \mathbf{D} \), where

\[
\mathbf{W} = \frac{1}{\sqrt{2}} \begin{pmatrix} \mathbf{U} & \mathbf{U} \end{pmatrix},
\]

\[
\mathbf{U} = \frac{1}{\sqrt{3}} \begin{pmatrix} 1 & 1 & 1 \\ 1 & 2 & x_1 \\ x_2 & x_3 & x_4 \end{pmatrix} \quad \text{with} \ x = \exp(i2\pi/3).
\]

The nonzero elements of \( \mathbf{D} \) consist of six \( 2 \times 2 \) matrices along the main diagonal and the bulk spin-wave energies are obtained from the solutions of the determinant conditions \( \det(\tilde{\mathbf{A}}(\phi) \pm \tau) = 0 \) for \( \phi = 0, \pm 2\pi/3 \) where we have defined \( \tilde{\mathbf{A}}(\phi) = \mathbf{A} + \mathbf{B} \exp(i\phi) + \mathbf{B}^\dagger \exp(-i\phi) \). Bulk spin-wave energies may be expressed as \( \omega_{n\mu}^2(k) \) with \( \phi = 0, \pm 2\pi/3 \), where

\[
\omega_{n\mu}^2(k) = [\Omega + \alpha \pm SJ'(k) + S\Phi(k, \phi)]^2(1 + \pm S1(1 + \sigma'))
\]

\[
\times J'(k_2) - 2SJ(1 + \sigma')\Phi(k, \phi)
\]

and

\[
\Phi(k, \phi) = 2\cos(\sqrt{3}k_\mu a/2)\cos(-k_\mu a/2 + \phi) + \cos(k_\mu a + \phi).
\]

**B. Easy-plane surface spin waves**

As in our previous work, calculations are first made for a semi-infinite system with a single (001) surface and for a thin film composed of \( N \) magnetic layers which are labeled using the layer index \( n = 1, \ldots, N \). The linearized equations of motion for the local spin raising and lowering operators at all the sites are formed and then transformed to a representation involving a two-dimensional wave vector \( \mathbf{k}_1 \) which runs parallel to the surfaces and a layer index \( n \). The spins located on the layers with an odd layer index \( n \) are taken to belong to sublattices \( A_1, B_1, \) and \( C_1 \) whereas those located on layers with an even layer index \( n \) belong to sublattices \( A_2, B_2, \) and \( C_2 \). For the \( l \)th site on sublattice \( A_1 \), the wavelike solution for the spin operator is written as

\[
S^\pm_{l_1} = S^\pm_{A_{l_1}}(k_1)\exp[i(k_1 \cdot \mathbf{r} - Et)],
\]

where the position vector \( \mathbf{r} = (x, y) \) and the amplitudes \( S^\pm_{A_{l_1}}(k_1) \) depend on the \( x \) coordinate through the layer index \( n \). Similar expressions are defined for sites on sublattices \( B_1, C_1, A_2, B_2, \) and \( C_2 \). For a semi-infinite system, the set of finite difference equations connecting the Fourier amplitudes may be expressed in supermatrix form as \( \mathbf{M} \mathbf{b} = 0 \), where \( \mathbf{M} \) is an \( \infty \times \infty \) block-tridagonal matrix with diagonal elements equal to \( \mathbf{A} \) (except for the leading element which is equal to \( A_1 \)) and off-diagonal elements \( \tau(0)/2 \).

Each element \( \mathbf{M} \) represents a \( 6 \times 6 \) matrix and \( \mathbf{A} \) is defined as in Eqs. (5) and (6). The elements of the matrix \( \mathbf{A}_1 \) are defined as in \( \mathbf{A} \) but with the substitution \( J \rightarrow J_1, D \rightarrow D_1, \) and \( J' \rightarrow J'/2 \). The differences between \( \mathbf{A}_1 \) and \( \mathbf{A} \) occur because of different exchange and anisotropy parameters for spins in the surface layer as well as the reduced number of interlayer neighbors for surface spins. The infinite column vector \( \mathbf{b} \) is written as \( \mathbf{b} = [\mathbf{b}_1, \mathbf{b}_2, \ldots]^T \), where \( \mathbf{b}_n \) is defined in Eq. (8) and \( n = 1, \ldots, \infty \). The equations are first partially decoupled by applying the transformation \( \mathbf{U}^{-1}\mathbf{M}\mathbf{U} \) to all of the elements of the supermatrix. The transformed equations may be written into three sets of finite difference equations as

\[
\tilde{\A}_1(\phi)\mathbf{X}_{1,\phi} + \frac{\tau(0)}{2}\mathbf{X}_{2,\phi} = 0 \quad (n = 1),
\]

\[
\frac{\tau(0)}{2}\mathbf{X}_{2,n,\phi} + \tilde{\A}_1(\phi)\mathbf{X}_{2,n+1,\phi} + \frac{\tau(0)}{2}\mathbf{X}_{2,n+2,\phi} = 0
\]

\[
(n = 2n + 1, n \geq 1),
\]

\[
\frac{\tau(0)}{2}\mathbf{X}_{2,n,\phi} + \tilde{\A}_1(\phi)\mathbf{X}_{2,n+1,\phi} + \frac{\tau(0)}{2}\mathbf{X}_{2,n+2,\phi} = 0
\]

\[
(n = 2n, n \geq 1),
\]

for \( \phi = 0, \pm 2\pi/3 \). The matrix \( \tilde{\A}_1(\phi) \) is defined as \( \tilde{\A}_1(\phi) = \tilde{\A}_1 + \tilde{\B}_1 \exp(i\phi) + \tilde{\B}_1\dagger \exp(-i\phi) \), whereas \( \tilde{\A}(\phi) \) is defined as in the bulk case. The column vectors in Eqs. (14)–(16) are written as \( \mathbf{X}_{n,\phi} = [X^+_n(\phi), X^-_n(\phi)]^T \) with elements

\[
X^+_n(\phi) = \frac{1}{\sqrt{3}}[S^+_A(\phi) + S^+_B(\phi) + S^+_C(\phi)]^T,
\]

where we omit the wave vector label \( k_1 \). Column vectors \( \mathbf{X}_{n,\phi} \) with an odd (even) layer index \( n \) involve linear combinations of the operator amplitudes on sublattice 1 (2) only. Equations (14) and (15) are used to eliminate the \( \mathbf{X}_{n,\phi} \) with \( n \) odd from the set of equations represented by Eq. (16). The resulting set of relations connecting \( \mathbf{X}_{n,\phi} \) with \( n \) even can be written in the matrix form

\[
\begin{pmatrix}
\mathbf{P}_{1,\phi} & \mathbf{Q}_{\phi} & 0 & 0 & 0 & \cdots \\
\mathbf{Q}_{\phi} & \mathbf{P}_{\phi} & \mathbf{Q}_{\phi} & 0 & 0 & \cdots \\
0 & \mathbf{Q}_{\phi} & \mathbf{P}_{\phi} & \mathbf{Q}_{\phi} & 0 & \cdots \\
\vdots & \ddots & \ddots & \ddots & \ddots & \ddots \\
\end{pmatrix}
\begin{pmatrix}
\mathbf{X}_{2,\phi} \\
\mathbf{X}_{2,\phi} \\
\mathbf{X}_{2,\phi} \\
\vdots
\end{pmatrix}
= 0,
\]

where

\[
\mathbf{P}_{1,\phi} = \tilde{\A}(\phi) - \frac{1}{4}\tau(0)[\tilde{\A}_1^{-1}(\phi) + \tilde{\A}^{-1}(\phi)]\tau(0),
\]

\[
\mathbf{P}_{\phi} = \tilde{\A}(\phi) - \frac{1}{2}\tau(0)\tilde{\A}^{-1}(\phi)\tau(0),
\]
\[ Q_\phi = -\frac{1}{4} \tilde{\pi}(0) \tilde{\Lambda}^{-1}(\phi) \tilde{\pi}(0). \]  

(19)

The three sets of equations given in Eq. (18) (for \( \phi = 0, \pm 2\pi/3 \)) couple the amplitudes on sublattice 2 only. The surface spin-wave frequencies can be numerically obtained following an analogous approach employed for ferromagnetically coupled layers.\(^{11}\)

For a thin film composed of \( N \) layers with (001) surfaces, the system of finite difference equations can be expressed in a supermatrix form as \( \mathbf{M} \mathbf{b} = 0 \) as for the semi-infinite case except that \( \mathbf{M} \) is now a \( 6N \times 6N \) block-tridiagonal matrix and the column vector is defined as \( \mathbf{b} = [\mathbf{b}_1, \ldots, \mathbf{b}_N]^T \). The first and the last elements on the main diagonal of the supermatrix \( \mathbf{M} \) are written as \( A_1 \) and \( A_N \), respectively. The elements of the matrix \( A_N \) describe the effects of the additional surface of the film and are defined as in \( A_1 \) for the semi-infinite case except that with the appropriate exchange and anisotropy parameters at the surface \( n = N \). The coupled systems of equations in the case of a thin film can be decoupled using the method outlined for a semi-infinite system. However, cases involving an even or an odd number of layers \( N \) must be dealt with separately. For a film containing an even number of layers, both surfaces belong to the same sublattice, whereas for a film with an even number of layers, the spins in the surface layers belong to different sublattices. The spin-wave excitation spectrum in a stacked triangular antiferromagnetic film is numerically obtained by solving the determinant condition \( \det \mathbf{M} = 0 \).

**C. Numerical results**

In Fig. 1, illustrative results are shown for the spin-wave frequency versus in-plane wave vector \( k_{xa} \) (with \( k_y = 0 \)) for a representative \( S = 1 \) quasi-1D planar triangular antiferromagnet where we set \( J = 1.0 \) GHz, \( J' = 100.0 \) GHz, and \( \sigma = \sigma' = 0.0 \). The dispersion relations in the top and bottom figures correspond to \( D' = 1.0 \) GHz and \( D' = 0.0 \), respectively. For clarity, the results for the dispersion relation are shown using an extended zone scheme.\(^{11,15}\) The surface exchange and anisotropy parameters are taken to be uniform throughout the system. The shaded areas represent all of the bulk solutions \( \omega_{k_x}(\phi) \) [see Eq. (11)] with \( 0 < k_x \alpha = \pi/2 \) and the three sections (from left to right) correspond to the bulk solutions with \( \phi = 0, 2\pi/3, -2\pi/3 \). The \( M \) point denotes the zone-edge wave vector \( \left(k_x, k_y\right) = \left(2\pi/3a, 0\right) \). For these parameter values, the top of the bulk regions extends to approximately 208 GHz and only the lower part of the effective bulk continuum is shown. The solid line represents a surface spin wave obtained from the solution of Eq. (18) for a semi-infinite system. This mode is characterized with an amplitude that decays into the film away from the surface. In the top figure, results are also shown for two lowest energy modes for thin films composed of 8 layers (dotted lines) and 16 layers (dashed lines). The gapless (Goldstone) excitation in each case is a uniform mode and reflects the fact that all of the spins can be simultaneously rotated about the crystal \( c \) axis without any cost in energy. In the bottom figure, the dotted, dashed, and solid lines represent two lowest energy excitations for a film composed of \( N = 9, 17, \) and 100 layers, respectively. The additional zero-energy mode corresponds to the simultaneous rotation of all the spins out of the \( ab \) plane. This operation does not cost energy in the limit of \( D' = 0.0 \). The dominant effect of the anisotropy is to open up a gap in the spectrum at the \( \Gamma \) point for the surface and bulk excitation. The gap energies for the bulk modes are obtained from Eq. (11) and are \( \omega_{k_x}(\pm 2\pi/3) = 5\left[D'\left(8J' + 9J\right)\right]^{1/2.} \) For most values of the wave vector, the surface mode obtained for a semi-infinite system is well separated from the bulk region. This contrasts with the behavior obtained for a quasi-1D easy-plane triangular antiferromagnet with ferromagnetic interlayer exchange coupling where the splitting between the surface branch and the bulk region is negligible for a system characterized with uniform exchange and anisotropy parameters.

The eigenvectors can be used to characterize the symmetry of the spin-wave modes in frustrated thin films and to describe the nature of the modes in terms of acoustical (in-phase) and optical (out-of-phase) precession. As an example, we consider the symmetry properties of the four lowest \( k_l = 0 \) energy modes for a quasi-1D antiferromagnetic film with an even number of layers. For \( N = 8 \), the four spin-wave modes have energies equal to \( \omega_1 = 0.0, \omega_2 = 20.7, \omega_3 = 29.3 \), and \( \omega_4 = 40.6 \) GHz (see Fig. 1). The Fourier amplitudes of
the transverse spin components \((S^x_{C,i}(k), S^y_{C,i}(k))\) (for \(i = A, B, C\)) within any particular layer \(n\) are numerically obtained from Eqs. (14)–(18). The amplitudes are defined with respect to the local coordinate systems defined by the transformation in Eq. (2). The mode \(\omega_1\) is characterized with \(S^x_{A,i}(k) = S^y_{A,i}(k) = S^z_{C,i}(k)\) and \(S^x_{B,i}(k) = S^y_{B,i}(k) = S^z_{C,i}(k)\) = 0. On any sublattice, the mode has a uniform amplitude throughout the thickness of the film and this mode is classified as an acoustic spin wave. The mode \(\omega_2\) is characterized with \((-S^x_{A,i}(k), S^y_{A,i}(k), S^z_{C,i}(k))\) and \((S^x_{B,i}(k), S^y_{B,i}(k), S^z_{C,i}(k))\) within any layer. On a given sublattice, the mode is antisymmetric with respect to the center of the film. For example, on sublattice \(A\), we have \((S^x_{A,i}(k), S^y_{A,i}(k)) = -(S^x_{A,i+1}(k), S^y_{A,i+1}(k))\). On any sublattice, the spin-wave mode \(\omega_3\) is symmetric about the center of the film and the amplitudes on the three sublattices satisfy \((S^x_{A,i}(k), S^y_{A,i}(k)) = (S^x_{B,i}(k), S^y_{B,i}(k)) = (S^x_{C,i}(k), S^y_{C,i}(k))\). Mode \(\omega_3\) antisymmetric with respect to the center of the film and the amplitudes on any layer satisfy \(S^x_{A,i}(k) = S^x_{B,i}(k) = S^x_{C,i}(k)\) and \(S^y_{A,i}(k) = S^y_{B,i}(k) = S^y_{C,i}(k)\). The higher energy modes \(\omega_4\), \(\omega_5\), and \(\omega_6\) involve some out-of-phase oscillations and are classified as optical spin waves. The relationships between the Fourier amplitudes for the four lowest \(k\) modes hold for any film composed of an even number of layers. Similar relationships can be obtained for a film composed of an odd number of layers.

The spin-wave dispersion relation in these frustrated films shows interesting characteristics. The dispersion relation in films composed of an odd number of layers is qualitatively similar to that with ferromagnetic interlayer exchange. This can be explained by the fact that for an odd number of layers, both surfaces belong to the same sublattice and each individual chain of spins will possess a net moment. For thin films with an even number of layers \(N\), the system is composed of antiferromagnetically coupled chains that possess no net moment because each chain of spins has an equal number of sites from sublattices 1 and 2. As the number of layers in the film increases, this effect becomes less important and the two lowest branches eventually become degenerate and equal to the results obtained for the semi-infinite system. The two modes correspond to localized excitations at each surface. The crossing point of the two low-energy branches in Fig. 1 (top) depends on the assumed parameters of the model.

In Fig. 2, results are shown for the surface and bulk spin-wave energies versus in-plane wave vector \(k_x, a\) for a semi-infinite quasi-2D system with \(J = 100.0\) GHz, \(J' = 1.0\) GHz, \(D' = 1.0\) GHz (with \(D_1 = D\)), and \(\sigma = \sigma' = 0.0\). The bulk spin waves form a very narrow continuum (shaded areas) in quasi-2D systems because of the weak dependence on the third wave vector component \(k_z\). Also shown are the substantial effects of a modified surface exchange coupling \(J_1\) on the surface spin waves. Splitting of the surface branch away from the bulk region can also be obtained by assuming a modified value for the surface anisotropy \(D_1\) and compared to the bulk value. The energy gap at the zone center \(\Gamma\) for the bulk modes \(\omega_n(\pm 2\pi/3)\) is due to the anisotropy and vanishes when \(D' = 0\).

FIG. 2. Surface and bulk spin-wave energy versus in-plane wave vector \(k_x, a\) for a semi-infinite quasi-2D system with \(J = 100.0\) GHz, \(J' = 1.0\) GHz, \(D' = 1.0\) GHz (with \(D_1 = D\)), and \(\sigma = \sigma' = 0.0\). The solid line appearing just below the bulk region (shaded area) corresponds to a case with uniform interlayer coupling \(J = J_1\), whereas the dotted and dashed lines are obtained with \(J_1 = 0.5J\) and \(J_1 = 0.75J\), respectively.

III. EASY-AXIS ANISOTROPY

A. Easy-axis bulk spin-wave excitations

Consider now the triangular antiferromagnet characterized with a single-ion anisotropy whose easy axis is along the crystallographic \(c\) axis. It is instructive to first carry out the calculations for the bulk spin-wave dispersion. We follow a method similar to the one described previously for the case of easy-plane anisotropy. For the bulk spin-wave calculation, the system can again be divided into the six sublattices as depicted schematically in Fig. 3(a). It is worth noting that the ground state spin configuration can be rotated about the \(c\) axis by any arbitrary angle and here we assume that all of the spins have their equilibrium positions in the \(ac\) plane.
out of generality, we set the equilibrium direction for spins on sublattice $A_1$ along the $c$ axis. The equilibrium configurations for spins on sublattices $B_1, C_1, A_2, B_2,$ and $C_2$ can be obtained using a local field method.\textsuperscript{16–18} The main effect of the easy-axis anisotropy on the equilibrium configuration is that the spins now lie in the $a$ plane containing the $c$ axis and a deformation of the 120° structure giving $\theta > 120^\circ$ as shown in Fig. 3(b).

The Hamiltonian in Eq. (1) is transformed to a local coordinate system such that the direction of the local $z$ axis is in the direction of the equilibrium spin alignment. The transformations for the sites on the six sublattices are written as

$$(S_i^x, S_i^y, S_i^z) \rightarrow [(-1)^{\mu+1}(S_i^x \cos \theta_i - S_i^y \sin \theta_i), S_i^y, (-1)^{\mu+1} \times (S_i^x \sin \theta_i + S_i^y \cos \theta_i)],$$

with $i=1, m_\mu, n_\mu, \theta_\mu = 0, \theta_n = \theta$, and $\theta_m = -\theta$, with $\mu=1, 2$. The stability condition requires that the coefficients of the local transverse spin components $S_i^x$ and $S_i^y$ vanish for all sites and leads to the bulk canting angle which is given by

$$\cos \theta = -\frac{(1 + \alpha)}{2[1 + \alpha^2 - \alpha' J'/3J + D'/3J]},$$

Equation (21) is a generalization of the results obtained in Refs. 16 and 19 which treat the cases of either easy-axis anisotropy or exchange anisotropy, but not both. In the absence of anisotropy ($\alpha = \alpha' = D' = 0$), the 120° structure is recovered, as expected. The linearized equations of motion for the various sites are calculated and then transformed to a wave vector representation. Twelve equations are required to obtain a closed set. The system of coupled equations may be expressed as $\mathbf{M} \nu = 0$, where the $12 \times 12$ matrix $\mathbf{M}$ has the same form as in Eq. (4) and the column vector $\nu$ is formally written as in Eq. (8). In this case, the elements of the supermatrix $\mathbf{M}$ are now given by

$$\mathbf{A} = \begin{pmatrix} \tilde{A} & \tilde{B} & \tilde{B}^* \\ \tilde{B}^* & \tilde{C} & \tilde{D} \\ \tilde{B} & \tilde{D}^* & \tilde{C} \end{pmatrix}, \quad \tau = \begin{pmatrix} \tau(k_1) \ 0 \ 0 \\ 0 \ \lambda(k_1) \ 0 \\ 0 \ 0 \ \lambda(k_2) \end{pmatrix},$$

where

$$\tilde{A} = \begin{pmatrix} E + \Omega & 0 \\ 0 & E - \Omega \end{pmatrix}, \quad \tilde{B} = -SJ(k_1)/2 \begin{pmatrix} c_1^+ & -c_1^- \\ -c_1^- & c_1^+ \end{pmatrix},$$

$$\tilde{C} = \begin{pmatrix} E + \alpha & \delta \\ -\delta & E - \alpha \end{pmatrix}, \quad \tilde{D} = -SJ(k_1)/2 \begin{pmatrix} c_2^+ & -c_2^- \\ -c_2^- & c_2^+ \end{pmatrix},$$

$$\tau(k_1) = SJ'(k_1) \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix},$$

$$\lambda(k_2) = SJ'(k_2)/2 \begin{pmatrix} \alpha' \sin^2 \theta & 2 + \alpha' \sin^2 \theta \\ -2 + \alpha' \sin^2 \theta & -\alpha' \sin^2 \theta \end{pmatrix},$$

with matrix elements defined as

$$\mathbf{M} = 2S(1 + \alpha \cos J'(0) + 2SD' - S(1 + \alpha')J'(0),$$

$$\alpha = S[1 + \alpha \cos \theta + \cos^2 \theta - \sin^2 \theta ]J'(0)$$

$$- S(1 - 3 \cos^2 \theta)D' - S(1 + \alpha' \cos^2 \theta)J'(0),$$

$$\delta = -SD' \sin^2 \theta,$$

$$c_1^+ = \cos \theta \pm 1,$$

$$c_2^+ = \cos^2 \theta - (1 + \alpha) \sin^2 \theta \pm 1,$$

$$D' = [1 - (2S)^{-1}]D.$$}

B. Spin waves in thin films

The spin-wave excitations in thin films composed of $N$ magnetic layers with (001) surfaces may be investigated by extending the method used for bulk spin waves. The calculations for a thin film with easy-axis anisotropy are algebraically more complicated than those for a film with easy-plane anisotropy because the system can no longer be described in terms of a six sublattice model. However, within any particular layer of the film, the system is still divided into three interpenetrating sublattices. Without loss of generality, the equilibrium direction for the surface layer spins $n=1$ on sublattice $A$ is set along the $c$ axis and the equilibrium direction for all of the other spins is obtained by minimizing the classical energy of the system. Sites on sublattice $A$ with an odd layer index $n$ also have their equilibrium directions along the $c$ axis, whereas sublattice A sites with an even layer index are antiparallel because of the antiferromagnetic interlayer exchange coupling. The canting angles for sites on sublattices $B$ and $C$ in layer $n$ are written as $\theta_{B,n} = \theta_{C,n} = \theta_n$. A schematic diagram of the system is illustrated in Fig. 4(a) along with the local set of axes for each sublattice. The 3N coordinate matrices may be written as

$$(S_i^x, S_i^y, S_i^z) \rightarrow [(1)^{n+1}(S_i^x \cos \theta_i - S_i^y \sin \theta_i), S_i^y, (-1)^{n+1} \times (S_i^x \sin \theta_i + S_i^y \cos \theta_i)\tau],$$

with $i=1, m_n, n_n$ with $n=1, \ldots, N$, and we define $\theta_0 = 0, \theta_{m_n} = \theta_n$, and $\theta_{n_n} = -\theta_n$. The Hamiltonian in Eq. (1) is again written with respect to the local coordinate systems, and the stability conditions for the spins on sublattice B or C are now given by

$$3J_n[\sin \theta_n + (1 + \alpha/2)\sin 2\theta_n] + D_n' \sin 2\theta_n$$

$$+ J'\{\sin \theta_{n-1} \cos \theta_n - (1 + \alpha') \cos \theta_{n-1} \sin \theta_n\}[1 - \delta_{n,n}],$$

$$\times [\sin \theta_{n+1} \cos \theta_n - (1 + \alpha') \cos \theta_{n+1} \sin \theta_n][1 - \delta_{n,n}],$$

$$= 0 \quad \text{for } n=1, \ldots, N.$$
The surface effects are taken into account through the
Kronecker $\delta$ in Eq. (26) and modified values of the exchange
and single-ion anisotropy at the surfaces of the film are
allowed. The set of $N$ nonlinear equations can be solved using
a numerical self-consistent approach. However, there are
a few limiting cases that can be solved analytically. If we
neglect the anisotropy terms ($\sigma = \sigma' = D_n = 0$), we recover the
120° structure in any particular layer, as expected. In the
limit of $J' = 0$, the layers are independent from one another
and the canting angle within any layer will be given by Eq.
(21). When the exchange and anisotropy surface parameters
are set equal to their bulk values, it is easily verified that the
canting angles for sites on sublattices $B$ and $C$ are layer
independent and again given by Eq. (21). For all other cases,
a numerical solution is required.

The spin-wave energies in thin films are obtained by
forming $6N$ linearized coupled equations of motion which
may be expressed as $\mathbf{M}\mathbf{b} = 0$, where $\mathbf{M}$ is a block-tridiagonal
matrix, with diagonal elements defined as $A_n$ and off-
diagonal elements equal to $\tau_{n+1}$ (with $i=1,N-1$), and $\mathbf{b} = [b_1, \ldots, b_{3N}]^T$. The elements of the matrices $A_n$ and $\tau_{n,n'}$
(defined in the Appendix) are similar to those in the bulk
case in Eqs. (22)–(24), where the main differences are due to
the layer-dependent canting angles for spins on sublattices $B$
and $C$. Spin-wave excitations are numerically obtained from
the solutions of $\det \mathbf{M} = 0$.

C. Numerical results

In all the cases studied, the spins on sublattice $A$ form
antiferromagnetic chains with ordering along the $c$ axis. The
canting angle is layer dependent only if the surface exchange
or anisotropy parameters are allowed to differ from their corre-
 respond ing bulk values. In quasi-1D systems, the strong in-
terlayer coupling tends to restore the antiferromagnetic order
in the spin chains on sublattices $B$ and $C$. For quasi-2D sys-
tems, the perturbation of the canting angle in cases with
modified surface parameters is localized to a small number
of layers (typically $\sim 2$) away from the surface.

An example of the spin-wave dispersion relation for a
quasi-1D system with easy-axis anisotropy is illustrated in
Fig. 5. The shaded area corresponds to the low-energy bulk
spin waves. The solid lines represent the six lowest energy
modes for a uniform film composed of $N=20$ layers. The
parameter values for the representative quasi-1D system are
$J = 1.0$ GHz, $J' = 0.0$ GHz, $D' = 1.0$ GHz, and $\sigma = \sigma' = 0.0$. All of the spin-waves modes obtained from the determinantal
condition $\det \mathbf{M} = 0$ are degenerate in magnitude and only the
positive solutions are shown. A Goldstone mode also exists
in bulk and thin films characterized with easy-axis aniso-
tropy because of the zero energy cost associated with rotat-
ing the spin configurations in Figs. 3 and 4 about the crystal
c axis.

The operator equation of motion method used in the
previous sections may be easily extended to calculate Green’s
functions which can be used to discuss the spectral intensi-
ities of the spin-wave modes predicted in this work. In order
to further characterize the low-energy spin-wave modes in
thin films with easy-axis anisotropy, the mean-squared am-
plitude of the spin precession, defined for sublattice $i$
($=A, B, C$) as $Q_i(k_{ix},n) = (S_{i,n}^z)^2$, is considered. For
simplicity, we outline the calculation of $Q_i(k_{ix},n)$. This real
quantity can be written in terms of equal time ($t=t'$) trans-
verse spin-correlation functions as

FIG. 4. (a) Schematic view of the spin configuration for a
stacked triangular antiferromagnet with easy-axis anisotropy and a
(001) surface as viewed along the chains. (b) Local coordinate sys-
tem for spins in layer $n$, where $n$ is an odd layer index.

FIG. 5. Spin-wave energy versus in-plane wave vector $k_{ix}$ (with
$k_{iy} = 0$) for an $S = 1$ quasi-1D system with easy-axis anisotropy.
The parameters are $J = 100.0$ GHz, $J' = 1.0$ GHz, $D' = 1.0$ GHz, and $\sigma = \sigma' = 0.0$. The exchange and anisotropy parameters at the surfaces
of the film are set equal to their corresponding bulk values. The
shaded area corresponds to the lower edge of the bulk continuum
and the solid lines correspond to the six lowest energy modes ob-
tained for a thin film composed of $N=20$ layers. The six lowest
energy branches for the film are labeled $\omega_1, \ldots, \omega_6$. 
\[ Q^A(k_i,n) = \frac{1}{2} \left( \langle S_{A,n}^+ (t) S_{A,n}^- (t) \rangle_{k_i} + \langle S_{A,n}^- (t) S_{A,n}^+ (t) \rangle_{k_i} \right). \]  

The spectral representation \[ \xi_n(k_i,E) \] of the transverse spin-correlation function \[ \langle S_{A,n}^+ (t) S_{A,n}^- (t') \rangle_{k_i} \] is defined as

\[ \langle S_{A,n}^+ (t) S_{A,n}^- (t') \rangle_{k_i} = \int_{-\infty}^{\infty} \xi_n(k_i,E) \exp(-iE(t-t')) dE \]  

is obtained from the imaginary part of the Green function \[ G^A_n(k_i,E) = \langle S_{A,n}^+ (k_i) S_{A,n}^- (k_i) \rangle_E \] using the fluctuation dissipation theorem. The spectral function of the other correlation function \[ \langle S_{A,n}^+ (t) S_{A,n}^- (t') \rangle_{k_i} \] may also be deduced from \[ G^A_n(k_i,E) \]. Some examples of the application of the Green function equation of motion method to magnetic thin films with a different ordering of the spins are given in Refs. 13 and 21–23. In the low-temperature limit \( T \ll T_C \), the solution to the Green function \[ G^A_n(k_i,E) \] may be written as

\[ G^A_n(k_i,E) = \frac{S}{\pi \det M} \left[ \text{adj} M \right]_{-5,0,-5} \text{ for } n = 1, ..., N, \]  

where \[ \left[ \text{adj} M \right]_{n,n} \] denotes the nth diagonal element of the adjoint of the matrix \( M \). Other diagonal elements of the adjoint of \( M \) are used to obtain the Green functions \( G^B_n(k_i,E) \) and \( G^C_n(k_i,E) \). The spin-wave energies obtained from \[ \det M = 0 \] will correspond to the poles of the Green functions. The mean-squared amplitude \[ Q^A(k_i,n) \] (or integrated intensities) of a particular spin-wave mode is estimated by evaluating the area under the peak of the imaginary parts of \[ G^A_n(k_i,E + i\epsilon) \], where the real and positive quantity \( \epsilon \) is introduced phenomenologically to model an intrinsic damping or reciprocal lifetime.

Figure 6 shows the mean-squared amplitudes as a function of the layer index corresponding to some of the \[ k_i = (2\pi/3a,0) \] spin-wave modes shown in Fig. 5. The mean-squared amplitudes for sublattices B and C within any particular layer are equal. Numerical results show that the spin-wave modes \( \omega_1, ..., \omega_A \) contribute mostly to the spin fluctuations on sublattices B or C compared with modes \( \omega_1, ..., \omega_B \). These higher energy modes are characterized with amplitudes that vary in a wavelike fashion across the thickness of the film on sublattices B and C. However, numerical results show that the spin-wave modes \( \omega_B \) and \( \omega_C \) are localized on sublattice A sites that are near the surfaces of the film. Higher energy modes are characterized with amplitudes that vary in a wavelike fashion on all three sublattices.

![Figure 6](image-url)

**FIG. 6.** Mean-squared amplitudes \( Q^B(k_i,n) = Q^B(k_i,n) \) plotted against the layer index \( n \) for some of the low-energy \( k_i = (2\pi/3a,0) \) modes defined in Fig. 5 for a quasi-1D film composed of \( N = 20 \) layers. The amplitudes \( Q^B(k_i,n) \) of the spin-wave modes \( \omega_1 \) and \( \omega_2 \) (not shown) are qualitatively similar to those of \( \omega_1 \). All of the results are symmetric with respect to the center of the film because of the symmetric choice of anisotropy and exchange parameters at the surfaces. The lines are guides for the eye. The closed symbols define the amplitudes on even numbered layers, whereas the open symbols are used for the odd numbered layers.

Figure 7 shows illustrative results for an \( S = 1 \) quasi-2D film composed of \( N = 20 \) layers where we set \( D' = -10 \) GHz, \( J' = 1.0 \) GHz, \( J = 100.0 \) GHz, and \( \sigma = \sigma' = 0.0 \). Exchange and anisotropy parameters at the surfaces are set equal to their corresponding bulk values. For each value of the in-plane wave vector \( k_i \), the solution of \( \det M = 0 \) yields a total of \( 6N \) spin-wave modes. All of the solutions are degenerate in magnitude and the dispersion relation (showing positive solutions only) is illustrated in Fig. 7(a). The modes are split into three distinct groups each containing a total of 20 branches. Qualitatively similar results (not shown) are obtained for a quasi-2D system with easy-plane anisotropy. The main effect of the easy-axis anisotropy is to remove the degeneracies.
between some of the spin modes. This is a consequence of the reduced symmetry in systems characterized with easy-axis anisotropy compared with systems with easy-plane anisotropy. Discussions of the symmetry properties of the bulk spin-wave modes can be found in Ref. 19. The lowest energy branch in each group is labeled $\omega_1$, $\omega_2$, and $\omega_3$ refer to the lowest energy branches in each group. The inset focuses on the small wave vector region and shows the splitting of the $\omega_3$ branch from the other modes in the group. (b) Sublattice $A$ spectral representation $-\text{Im} \{G_{\text{sub}}^{A}(\mathbf{k}, E+i\epsilon)\}$ with $\mathbf{k}_0=(2\pi/3a,0)$ and $\epsilon=0.01$ GHz versus frequency in layers $n=1$ (solid line) and $n=3$ (dashed line). (c) The amplitude $Q^{A}(k, n)$ (integrated intensity) of the zone-edge surface modes $\omega_1$, $\omega_2$, and $\omega_3$ as a function of the layer index. Results are symmetric about the center of the film and only results from the first four layers are shown. Lines are guides for the eye and have no physical meaning. (d) The mean-squared amplitude $Q^{B}(k, n)$ for the bulk excitation $\omega_B$ defined in (b) versus layer index. Open and close symbols are used for odd and even layers, respectively.

FIG. 7. (a) Spin-wave dispersion relation for an $S=1$ easy-axis quasi-2D film composed of 20 layers with uniform parameters where we set $D'=-10$ GHz, $J'=1.0$ GHz, $J=100.0$ GHz, and $\sigma=\sigma'=0.0$. The spin-wave modes are split into three groups each containing 20 branches. The labels $\omega_1$, $\omega_2$, and $\omega_3$ refer to the lowest energy branches in each group. The inset focuses on the small wave vector region and shows the splitting of the $\omega_3$ branch from the other modes in the group. (b) Sublattice $A$ spectral representation $-\text{Im} \{G_{\text{sub}}^{A}(\mathbf{k}, E+i\epsilon)\}$ with $\mathbf{k}_0=(2\pi/3a,0)$ and $\epsilon=0.01$ GHz versus frequency in layers $n=1$ (solid line) and $n=3$ (dashed line). (c) The amplitude $Q^{A}(k, n)$ (integrated intensity) of the zone-edge surface modes $\omega_1$, $\omega_2$, and $\omega_3$ as a function of the layer index. Results are symmetric about the center of the film and only results from the first four layers are shown. Lines are guides for the eye and have no physical meaning. (d) The mean-squared amplitude $Q^{B}(k, n)$ for the bulk excitation $\omega_B$ defined in (b) versus layer index. Open and close symbols are used for odd and even layers, respectively.
The results presented above outline the application of an operator formalism to calculate surface and thin-film effects on the spin dynamics in AF-coupled geometrically frustrated triangular layers. This serves to extend our previously published work on F-coupled films to the physically more relevant case applicable to a large variety of materials. Through the use of illustrative numerical calculations, differences in linear spin excitations for bulk, semi-infinite, and thin-film systems having either easy-plane or easy-axis anisotropy and quasi-1D or quasi-2D exchange couplings are highlighted.

A number of features of these results are of interest. In contrast with the case of F-coupled films, bulk and surface mode excitations remain well separated in the case of AF-coupled layers even in the case where surface exchange and anisotropy parameters are identical to bulk values. Differences also arise between these two types of interlayer couplings in the case of thin films. In the case of AF coupling, the results are dependent on whether there are an even or odd number of layers corresponding to having compensated or uncompensated total moments, respectively. Most of our knowledge regarding the sign of exchange interactions is based on rules which may not be entirely applicable to surface and thin-film environments. Examination of the spin excitation spectrum in view of these findings offers a possible tool to distinguish the type of interlayer coupling. The present work also features a calculation of mode amplitudes. The mode amplitudes exhibit a variety of dependences through the thickness of the thin films and if the system is quasi-1D or quasi-2D. Potential extensions of the present calculations might be to include dipolar effects that can be important at surfaces even in AF systems. This may require the utilization of purely numerical algorithms such as those based on classical equations of motion. Another interesting extension of the work would be the study of the static and dynamic magnetic properties of superlattice structures composed of different frustrated antiferromagnetic thin films, extending work done on unfrustrated superlattice structures. The magnetic properties of composite materials can be quite distinct from any individual constituent due to interface effects between different magnetic materials.

IV. CONCLUSIONS

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APPENDIX

The definitions of matrices $A_n$ and $\tau_{n,n'}$ for a stacked triangular antiferromagnetic thin film with easy-axis anisotropy are
\[ A_n = \begin{pmatrix} \bar{A}_n & \bar{B}_n & \bar{B}_n^* \\ \bar{B}_n & \bar{C}_n & \bar{D}_n \\ \bar{B}_n & \bar{D}_n & \bar{C}_n \end{pmatrix}, \quad \tau_{n,n'} = \begin{pmatrix} \tau_{n,n'} & 0 & 0 \\ 0 & \lambda_{n,n'} & 0 \\ 0 & 0 & \lambda_{n,n'} \end{pmatrix} \]

where

\[ \bar{A}_n = \begin{pmatrix} E + \Omega_n & 0 \\ 0 & E - \Omega_n \end{pmatrix}, \quad \bar{B}_n = -SJ(k) / 2 \begin{pmatrix} c_{1,n}^+ & c_{1,n}^- \\ c_{1,n}^- & c_{1,n}^+ \end{pmatrix}, \]

(A1)

and

\[ \lambda_{n,n'} = SJ'/2 \begin{pmatrix} \alpha' \sin \theta_n \sin \theta_{n'} & 2 \cos(\theta_n - \theta_{n'}) + \alpha' \sin \theta_n \sin \theta_{n'} \\ -2 \cos(\theta_n - \theta_{n'}) + \alpha' \sin \theta_n \sin \theta_{n'} & -\alpha' \sin \theta_n \sin \theta_{n'} \end{pmatrix} \]

(A3)

with matrix elements defined as

\[ \Omega_n = 2S(1 + \sigma)\cos \theta_n J(0) + 2SD' - S(1 + \sigma')J'[2 - \delta_{1,n} - \delta_{n,N}], \]

\[ \alpha_n = S[(1 + \sigma)(\cos \theta_n + \cos^2 \theta_n) - \sin^2 \theta_n J(0)] - S[1 - 3 \cos^2 \theta_n]D' - SJ'[\cos(\theta_n - \theta_{n+1}) + \sigma' \cos \theta_n \cos \theta_{n+1}](1 - \delta_{1,n}) \]

\[ + [\cos(\theta_n - \theta_{n+1}) + \sigma' \cos \theta_n \cos \theta_{n+1}](1 - \delta_{n,N}) \]

\[ \delta_n = -SD' \sin^2 \theta_n, \]

\[ c_{1,n}^+ = \cos \theta_n \pm 1, \]

\[ c_{2,n}^+ = \cos^2 \theta_n - (1 + \sigma)\sin^2 \theta_n \pm 1, \]

\[ D_n' = [1 - (2S)^{-1}]D_n. \]

(A4)

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