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Theory of surface and bulk excitations in ferromagnetic semiconductors

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The s-d (or s-f) interaction model is used to study the bulk and surface magnetic excitations of a semi-infinite ferromagnetic semiconductor. A formalism is established to obtain Green functions which provide the dynamic response of the system and from which the surface and bulk excitations and their dispersion relations are calculated. Results are deduced for both narrow-band and wide-band ferromagnetic semiconductors. The applicability to particular materials, such as the spinel-type ferromagnetic semiconductors is discussed.

I. INTRODUCTION

There has been considerable interest in the study of surface excitations in various magnetically ordered systems. It has been shown for insulators within the framework of the Heisenberg exchange model that in addition to the bulk spin-wave excitations there may be surface spin waves whose amplitudes decay exponentially away from the surface of the material toward the interior (e.g., see Refs. 1–3 for reviews).

Surface magnetic modes have also been treated extensively by macroscopic methods involving Maxwell's equations in cases where the effects of long-range dipole-dipole interactions prevail over the short-range exchange interactions. In the magnetostatic regime these surface modes were studied in detail for ferromagnetic slabs,[4] and this type of calculation has more recently been extended to infinite and semi-infinite superlattices of ferromagnetic and antiferromagnetic metals and semiconductors in various geometries.[5–8] The conditions for magnetostatic theory can often be realized to a good approximation in light-scattering experiments,[9–11] where excitations of very small wave vectors are probed.

A microscopic approach to the surface spin waves in itinerant electron systems has been recently addressed[12,13] in the context of the Hubbard model. In this case different approximations introduced for the surface have led to some differing predictions for the spin-wave spectrum of ferromagnetic metals.

In this paper we address the problem of obtaining the surface excitations in magnetic semiconductors where a clear distinction can be made between localized and itinerant spins (e.g., see Refs. 14–17 for reviews). We present here a microscopic calculation of the surface and bulk excitations in degenerate ferromagnetic semiconductors using the s-d (or s-f) interaction model which consists of a Heisenberg ferromagnet interacting with the itinerant spins of the conduction electrons via a contact-type s-d (or s-f) interaction. Some of the ferromagnetic semiconductors of interest include europium chalcogenides (such as EuO and EuS) and spinels (such as $CdCr_2S_4$, $CdCr_2Se_4$, and $HgCr_2Se_4$); others are listed in Ref. 15. In some cases aspects of their magnetic properties have been studied experimentally by light scattering, either by Brillouin scattering from low-frequency spin waves or by spin-dependent Raman scattering from optical phonons (e.g., see Refs. 11, 18, and 19, and references therein).

There have been some calculations[20–23] applying the s-d (or s-f) model to *infinitely extended* ferromagnetic semiconductors. In addition to the usual "acoustic" spin-wave branch[20] it has been shown[21–23] that there is a higher frequency (or "optical") spin-wave branch and the Stoner-like continuum of magnetic excitations. Estimates were given[21] for the frequencies and relative intensities for light scattering from the two spin-wave branches. In the present work we show that there may also be localized (or surface) spin waves in the case of semi-infinite ferromagnetic semiconductors. Low-energy surface states in semi-infinite ferromagnetic semiconductors have previously been considered in the magnetic-polaron regime.[24,25]

In Sec. II we introduce the Hamiltonian describing a ferromagnetic semiconductor and obtain general expressions for the magnetic Green functions within a matrix formalism. A general solution of the problem is rather difficult analytically and so we proceed to consider some special cases in more detail. In Sec. III we consider narrow-band semiconductors (such as the chromium spinels) with $W \ll IS$, where W is the conduction bandwidth, I is the contact interaction energy, and S is the spin of the localized electrons. Some numerical illustrations of the surface and bulk spin-wave spectrum are presented in this section for the limiting case of $W=0$. In Sec. IV we present some results for wide-band semiconductors (such as EuO) with $W \gg IS$ where we use linear response arguments to solve for the magnetic Green functions. In doing so the transverse spin susceptibility of the conduction electrons has to be evaluated and our results are compared with some previous calculations.[26,12] The general evaluation of the surface spin-wave modes in this regime can only be done by numerical means. In Sec. V we present some analytical results for the case of narrow-band ferromagnetic semiconductors

with a small but finite width. We present the conclusions in Sec. VI and also provide in here some discussion of the surface modifications to the Stoner continuum.

II. MATRIX FORMALISM FOR THE MAGNETIC GREEN FUNCTIONS

In this section we present calculations for obtaining the magnetic Green functions for a semi-infinite degenerate ferromagnetic semiconductor occupying the half-space $z \geq 0$ and described by the s-d (or s-f) interaction model. The full Hamiltonian of the system is expressed as the sum of three terms: a Heisenberg Hamiltonian $\mathcal{H}_M$ for the localized spins (of d or f type), a Hamiltonian $\mathcal{H}_E$ representing the kinetic and Zeeman energy of the conduction (s) electrons, and an s-d (or s-f) interaction Hamiltonian $\mathcal{H}_I$, where

$$\mathcal{H}_M = -\tfrac{1}{2}\sum_{i,j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j - g\mu_B H_0 \sum_i S_i^z , \tag{1}$$

$$\mathcal{H}_E = \sum_{i,j,\sigma} t_{ij} a_{i\sigma}^{\dagger} a_{j\sigma} - g_e\mu_B H_0 \sum_i s_i^z , \tag{2}$$

$$\mathcal{H}_I = -\sum_i I_i \mathbf{S}_i\cdot\mathbf{s}_i . \tag{3}$$

Here $\mathbf{S}_i$ and $\mathbf{S}_j$ are spin operators for the localized spins at sites i and j in the semi-infinite medium, J_{ij} is the Heisenberg exchange interaction, assumed isotropic, and H_0 is a static magnetic field applied in the z direction (the direction of static magnetization). Also, t_{ij} is a hopping term and I_i is a contact interaction energy. The spin operators $\mathbf{s}_i$ of the conduction electrons at site i can be expressed as $s_i^+ = a_{i+}^{\dagger} a_{i-}$, $s_i^z = (a_{i+}^{\dagger} a_{i+} - a_{i-}^{\dagger} a_{i-})/2$, where $a_{i\sigma}^{\dagger}$ and $a_{i\sigma}$ are, respectively, the creation and destruction operators for an electron at site i and having a spin index σ (with $\sigma = \pm 1$ corresponding to up and down projections of the electron spin with respect to z, the quantization axis). We have denoted the Landé g factors of the localized and conduction electrons as g and g_e, respectively. It has been found[15] that the s-d interaction may greatly modify g_e from its free-electron value of 2. We have neglected in Eq. (2) the direct Coulomb interaction between the itinerant electrons (as would be included in the Hubbard Hamiltonian). Instead, the itinerant electrons have an indirect interaction via their coupling to the localized spin system.

We now introduce the transverse magnetic retarded commutator Green functions for the localized spin operators, defined in the conventional way as[27]

$$\langle\langle S_i^+(t); S_j^-(0)\rangle\rangle = -i\theta(t)\langle [S_i^+(t), S_j^-(0)]\rangle . \tag{4}$$

By constructing the frequency Fourier transform $\langle\langle S_i^+; S_j^-\rangle\rangle_\omega$ from the standard Green-function equation of motion used in the random-phase approximation at $T \ll T_c$, we obtain the following equation:

$$\left[\hbar\omega - g\mu_B H_0 - I_i\langle s_i^z\rangle - S\sum_l J_{li}\right]\langle\langle S_i^+; S_j^-\rangle\rangle_\omega + S\sum_l J_{il}\langle\langle S_l^+; S_j^-\rangle\rangle_\omega = \frac{S}{\pi}\delta_{ij} - I_i S\langle\langle s_i^+; S_j^-\rangle\rangle_\omega . \tag{5}$$

Here S is the averaged spin projection of the localized electrons (assumed independent of the site at $T \ll T_c$). We exploit the translational invariance in the xy plane to define the Fourier transform

$$\langle\langle S_i^+; S_j^-\rangle\rangle_\omega = \frac{1}{N}\sum_{\mathbf{k}_\parallel} \exp[i\mathbf{k}_\parallel\cdot(\mathbf{r}_i - \mathbf{r}_j)] G_{m,n}(\mathbf{k}_\parallel,\omega) , \tag{6}$$

where $\mathbf{k}_\parallel = (k_x, k_y)$ is a two-dimensional wave vector parallel to the surface, and m and n are positive integers labeling the lattice planes (parallel to the surface) that contain sites i and j, respectively. Hence $n = 1$ is the surface layer, $n = 2$ the next layer, and so on. The normalization constant N in Eq. (6) denotes the number of sites in any of the lattice planes.

We now simplify Eq. (5) by assuming that the localized spins are arranged in a simple-cubic structure with a as the lattice parameter and with crystallographic axes parallel to the coordinate xyz axes. (Note that the coordination of the nonmagnetic ions in a compound semiconductor may generally be such as to make the overall symmetry noncubic.) We further assume for simplicity only nearest-neighbor exchange interactions and take J_{ij} to have the value J_s if both spins i and j are in the surface layer ($n = 1$) and the bulk value J otherwise. We similarly assume that the hopping term t_{ij} takes the value t_s if both the electrons at sites i and j are in the surface layer, and otherwise it takes the bulk value t. We also assume that the on-site contact interaction energy I_i takes the perturbed value I_s only at the surface layer, but otherwise it has its corresponding bulk value I. Finally we take, for generality, $\langle s_i^z\rangle$ to have the value $\langle s_s^z\rangle$ only at the surface layer and the bulk value of $\langle s^z\rangle$ in every other layer. We can now rewrite Eq. (5), after using the Fourier transform in Eq. (6), in terms of infinite-dimensional matrices as

$$(\underline{A}_0 + \underline{D}_0)\underline{G}(\mathbf{k}_\parallel,\omega) = -\frac{1}{\pi J}\underline{I}_0 + \frac{I}{J}\underline{R}\,\underline{G}'(\mathbf{k}_\parallel,\omega) , \tag{7}$$

where the elements of the matrix $\underline{G}(\mathbf{k}_\parallel,\omega)$ are the Green functions $G_{m,n}(\mathbf{k}_\parallel,\omega)$ defined in Eq. (6) and the elements of $\underline{G}'(\mathbf{k}_\parallel,\omega)$ are similarly defined wave-vector Fourier transforms of the Green functions $\langle\langle s_i^+; S_j^-\rangle\rangle_\omega$ for the conduction spin and localized spin operators. Also, $\underline{I}_0$ is the unit matrix, and $\underline{R}$ is equal to the unit matrix with the (1,1) element replaced by $\nu \equiv I_s/I$. The quantity $\underline{A}_0$ is a tridiagonal matrix

$$\underline{A}_0 = \begin{bmatrix} d_0 & -1 & 0 & \cdots & & \\ -1 & d_0 & -1 & 0 & \cdots & \\ 0 & -1 & d_0 & -1 & 0 & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \end{bmatrix} , \tag{8}$$

where

$$d_0=(g\mu_B H_0+I\langle s^z\rangle-\hbar\omega)/SJ+2+4[1-\gamma(\mathbf{k}_\parallel)]\ , \tag{9}$$

$$\gamma(\mathbf{k}_\parallel)=[\cos(k_x a)+\cos(k_y a)]/2\ . \tag{10}$$

$\underline{D}_0$ is a matrix whose only nonzero element is $(\underline{D}_0)_{11}=\delta_0$, where

$$\delta_0=-1-4[1-\gamma(\mathbf{k}_\parallel)](1-J_s/J) -(\langle s^z\rangle I/SJ)(1-I_s\langle s_s^z\rangle/I\langle s^z\rangle)\ . \tag{11}$$

When I and I_s are set equal to zero Eqs. (7)–(11) reduce to those for a semi-infinite Heisenberg ferromagnet, and the solution for $\underline{G}(\mathbf{k}_\parallel,\omega)$ can then be expressed analytically since the inverse of a matrix having the form of $\underline{A}_0$ is known analytically (e.g., see Ref. 28). In the present case of a ferromagnetic semiconductor, however, we need to set up another equation connecting the Green-function matrices $\underline{G}(\mathbf{k}_\parallel,\omega)$ and $\underline{G}'(\mathbf{k}_\parallel,\omega)$. This is in general complicated and we consider first some special cases.

III. THE NARROW-BAND LIMIT ($t=0$)

If the hopping term t is small enough (compared with I), the main effect of the conduction electrons is through the interaction term $\mathcal{H}_I$ of Eq. (3). In the case of $t=0$ it is straightforward to construct the equation of motion satisfied by the Green function $\underline{G}'(\mathbf{k}_\parallel,\omega)$ to show that it yields

$$\underline{G}'(\mathbf{k}_\parallel,\omega)=-\frac{I\langle s^z\rangle}{\hbar\omega-IS-g_e\mu_B H_0}\underline{R}'\underline{G}(\mathbf{k}_\parallel,\omega)\ . \tag{12}$$

Here $\underline{R}'$ is equal to the unit matrix with the (1,1) element replaced by ν', where we define

$$\nu'=-\frac{[I_s\langle s_s^z\rangle(\hbar\omega-IS-g_e\mu_B H_0)]}{[I\langle s^z\rangle(\hbar\omega-I_sS-g_e\mu_B H_0)]}\ . \tag{13}$$

From Eqs. (7) and (12) we obtain

$$(\underline{A}+\underline{D})\underline{G}(\mathbf{k}_\parallel,\omega)=-\frac{1}{\pi J}\underline{I}_0\ , \tag{14}$$

where $\underline{A}$ and $\underline{D}$ are defined similarly to the matrices $\underline{A}_0$ and $\underline{D}_0$, except that the quantities d_0 and δ_0 are replaced by

$$d=d_0+I^2\langle s^z\rangle/[J(\hbar\omega-IS-g_e\mu_B H_0)]\ , \tag{15}$$

$$\delta=\delta_0-I^2\langle s^z\rangle/[J(\hbar\omega-IS-g_e\mu_B H_0)] +I_s^2\langle s_s^z\rangle/[J(\hbar\omega-I_sS-g_e\mu_B H_0)]\ , \tag{16}$$

respectively. We can now write the formal solution for $\underline{G}(\mathbf{k}_\parallel,\omega)$ as

$$\underline{G}(\mathbf{k}_\parallel,\omega)=(-1/\pi J)(\underline{I}_0+\underline{A}^{-1}\underline{D})^{-1}\underline{A}^{-1}\ . \tag{17}$$

Expressions for $\underline{A}^{-1}$ can be written down by analogy with earlier work[28] on surface magnetic excitations. This involves introducing the complex variable ξ which satisfies $|\xi|\le 1$ and

$$\xi+\xi^{-1}=d\ . \tag{18}$$

The matrix elements of $\underline{A}^{-1}$ are then given by

$$(\underline{A}^{-1})_{mn}=(\xi^{m+n}-\xi^{|m-n|})/(\xi-\xi^{-1})\ . \tag{19}$$

The bulk spin waves correspond to values of the frequency ω such that $|\xi|=1$ whereas the localization condition for the existence of surface spin waves is $|\xi|<1$. For the case of bulk spin waves we can write ξ as $\exp(ik_z a)$, where k_z is a real wave-vector component perpendicular to the surface, and the bulk spin-wave frequencies are the solutions of

$$\hbar\omega-g\mu_B H_0-2SJ[3-2\gamma(\mathbf{k}_\parallel)-\cos(k_z a)]-I\langle s^z\rangle -I^2\langle s^z\rangle S/(\hbar\omega-IS-g_e\mu_B H_0)=0\ . \tag{20}$$

This is consistent, as expected, with the dispersion relation for a spin wave with three-dimensional wave vector $\mathbf{k}=(\mathbf{k}_\parallel,k_z)$ in an *infinite* ferromagnetic semiconductor[22] in the limit of $t=0$. There are two branches to the bulk spin-wave spectrum, and if $I\gg J$ (which is typically the case) they are well separated in frequency.

The surface spin waves in the semi-infinite ferromagnetic semiconductor correspond to real values of ξ with $|\xi|<1$. The spin-wave frequencies can be obtained directly by noting from Eq. (17) that $\underline{G}(\mathbf{k}_\parallel,\omega)$ has poles corresponding to the condition $\det(\underline{I}_0+\underline{A}^{-1}\underline{D})=0$. This gives $\xi=-1/\delta$, where δ is defined in Eq. (16), and eventually it leads to the following expression satisfied by the surface spin-wave frequencies:

$$\hbar\omega-g\mu_B H_0-2SJ-4SJ[1-\gamma(\mathbf{k}_\parallel)]-SJ(\delta+\delta^{-1}) -I\langle s^z\rangle-I^2\langle s^z\rangle S/(\hbar\omega-IS-g_e\mu_B H_0)=0\ , \tag{21}$$

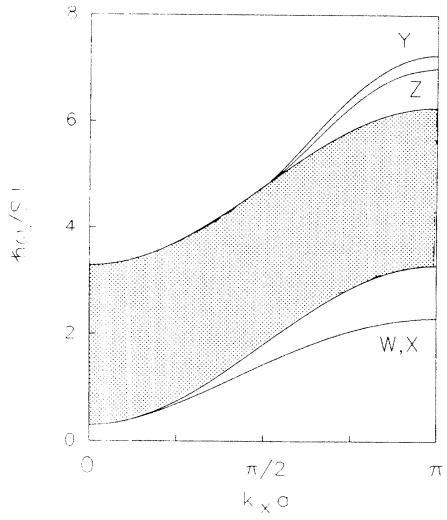

FIG. 1. The spin-wave frequencies (in units of $SJ/\hbar$) plotted against $k_x a$ for a semi-infinite narrow-band semiconductor (with $t=0$) in the low-frequency region. The parameter values are $I/J=200$, $S=\frac{3}{2}$, $\langle s^z\rangle=\frac{1}{2}$, $g\mu_B H_0/SJ=0.3$, and $g_e=g$. The bulk spin-wave region is shaded, and the labeling of the surface spin-wave branches corresponds to *W*, $J_s/J=0.5$, $I_s/I=1$; *X*, $J_s/J=0.5$, $I_s/I=0.2$; *Y*, $J_s/J=2$, $I_s/I=1$; *Z*, $J_s/J=2$, $I_s/I=0.2$.

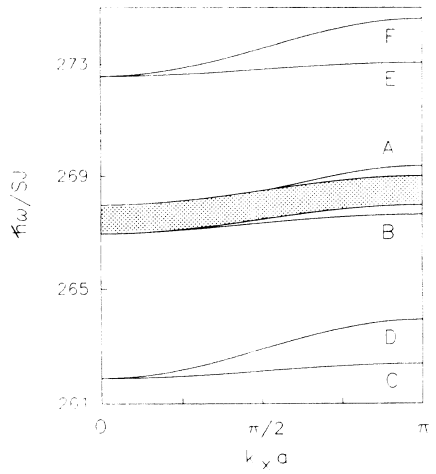

FIG. 2. As in Fig. 1, but in the high-frequency region. In this case the labeling of the surface spin-wave branches corresponds to *A*, $J_s/J=2$, $I_s/I=1$; *B*, $J_s/J=0.5$, $I_s/I=1$; *C*, $J_s/J=0.5$, $I_s/I=0.98$; *D*, $J_s/J=2$, $I_s/I=0.98$; *E*, $J_s/J=0.5$, $I_s/I=1.02$; *F*, $J_s/J=2$, $I_s/I=1.02$.

provided the existence condition $|\delta|>1$ is satisfied. We note from Eq. (16) that δ is ω dependent in the general case of $I_s \neq I$ and/or $\langle s_s^z\rangle \neq \langle s^z\rangle$, and Eqs. (16) and (21) then have to be solved self-consistently in order to obtain the surface spin-wave solutions. For simplicity we shall henceforth assume $\langle s_s^z\rangle$ to be the same as the bulk parameter $\langle s^z\rangle$, although the general case is included in our formalism.

Some numerical examples to illustrate the results for the spin waves are shown in Figs. 1 and 2 for the low-frequency and high-frequency excitations, respectively. In each case $\hbar\omega/SJ$ is plotted against $\mathbf{k}_\parallel a$ for various combinations of the exchange and *s*-*d* interaction parameters J_s/J and I_s/I. We have taken approximate values of the bulk parameters I and J appropriate to the $S=\frac{3}{2}$ ferromagnet $CdCr_2Se_4$, which is usually considered to be a narrow-band material.[15] The bulk spin waves each appear as a continuum in each of these plots, with upper and lower edges corresponding to $k_z=\pi/a$ and $k_z=0$, respectively, while the surface spin waves appear as discrete branches that may be above or below the continuum depending on the ratios J_s/J and I_s/I. We note from Fig. 1 that, for the low-frequency excitations, the surface spin-wave frequencies depend strongly on J_s/J but only weakly on I_s/I. From Fig. 2 for the high-frequency excitations it is apparent that the reverse applies.

IV. SOLUTIONS IN THE GENERAL CASE ($t\neq 0$)

The evaluation of the Green function $\underline{G}'(\mathbf{k}_\parallel,\omega)$ for general values of t is rather complicated. However, we can write down approximate solutions using linear-response theory to take account of the interaction term $\mathcal{H}_I$ in the Hamiltonian. This approach should be valid for the wide-band semiconductors (where $W \gg IS$), which were excluded from the analysis in Sec. III. Note that the bandwidth W is related to the bulk hopping parameter by $W=12t$ for a simple cubic lattice. Also, for the limiting case of $W\to 0$, we will see in the next section that the results obtained using these linear-response arguments correctly reduce to those described in Sec. III. We reexpress $\mathcal{H}_I$ of Eq. (3) as a sum of a static part $\mathcal{H}_I^S$ and a dynamic (or spin-flip) part $\mathcal{H}_I^D$. The static part can be incorporated into the Hamiltonian describing the conduction electrons and we can replace Eqs. (2) and (3) with

$$\mathcal{H}'_E=\mathcal{H}_E+\mathcal{H}_I^S$$
$$=\sum_{i,j,\sigma} t_{ij}a_{i\sigma}^\dagger a_{j\sigma}-\tfrac{1}{2}\sum_{i,\sigma}(I_iS+g_e\mu_BH_0)\sigma a_{i\sigma}^\dagger a_{i\sigma}\ , \tag{22}$$

$$\mathcal{H}'_I=\mathcal{H}_I^D=-\tfrac{1}{2}\sum_i I_i(S_i^+s_i^-+S_i^-s_i^+)\ . \tag{23}$$

In the regime where $\mathcal{H}'_I$ is a small perturbation to the Hamiltonian $\mathcal{H}'_E$ describing the conduction electrons we can apply linear-response theory[29] to describe the time evolution of the operator s_i^+ as

$$s_i^+(t)=-\tfrac{1}{2}\sum_j\int_{-\infty}^{\infty} I_j\langle\langle s_i^+(t);s_j^-(t')\rangle\rangle_0 S_j^+(t')dt'\ , \tag{24}$$

where the retarded Green function $\langle\langle s_i^+(t);s_j^-(t')\rangle\rangle_0$ is to be evaluated with respect to the Hamiltonian $\mathcal{H}'_E$ alone.

Since $\mathcal{H}'_E$ does not contain any spin-flip terms we can write the frequency Fourier transform $g_{ij}^0(\omega)$ of $\langle\langle s_i^+(t);s_j^-(t')\rangle\rangle_0$ in terms of single-particle spectral functions as (e.g., see Ref. 26)

$$g_{ij}^0(\omega)=\frac{1}{2\pi}\int d\omega_1\int d\omega_2\left[\frac{f(\omega_2)-f(\omega_1)}{\omega-\omega_1+\omega_2+i\varepsilon}\right]\times\left[\frac{J_{ji-}^0(\omega_1)J_{ij+}^0(\omega_2)}{f(\omega_1)f(\omega_2)}\right], \tag{25}$$

where ε is a positive infinitesimal, $f(\omega)$ are the Fermi distribution functions, and $J_{ij\sigma}^0(\omega)$ are the single-particle spectral functions for the electrons with spin projection σ ($\sigma=\pm1$). These spectral functions are directly related (through the fluctuation-dissipation theorem) to the single-particle Green functions. On introducing two-dimensional wave-vector Fourier transforms as before we obtain from Eq. (25)

$$g_{mn}^0(\mathbf{k}_\parallel,\omega)=\frac{2}{\pi N}\sum_{\mathbf{q}_\parallel}\int d\omega_1\int d\omega_2\left[\frac{f(\omega_2)-f(\omega_1)}{\omega-\omega_1+\omega_2+i\varepsilon}\right]\times\rho_{mn-}^0(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega_1)\times\rho_{nm+}^0(\mathbf{q}_\parallel,\omega_2)\ , \tag{26}$$

where m and n are the layer indices corresponding to sites i and j. The spectral weights $\rho_{mn\sigma}^0(\mathbf{k}_\parallel,\omega)$ are simply the imaginary parts of the corresponding one-electron Green functions $g_{mn\sigma}^0(\mathbf{k}_\parallel,\omega)$. These are evaluated by obtaining their standard equations of motion using $\mathcal{H}'_E$, reexpressing the resulting coupled equations in terms of a tridiagonal matrix, and then solving by the techniques described in the previous section. The results are

$$g^0_{mn\sigma}(\mathbf{k}_\parallel,\omega)=\frac{1}{2\pi t(x_\sigma-x_\sigma^{-1})}\left[x_\sigma^{m+n}\left[\frac{1+x_\sigma^{-1}\Delta_\sigma}{1+x_\sigma\Delta_\sigma}\right]-x_\sigma^{|m-n|}\right], \quad (27)$$

where x_σ is a complex variable with $|x_\sigma|\leq 1$ and defined as

$$x_\sigma+x_\sigma^{-1}=\hbar\omega/t-4\gamma(\mathbf{k}_\parallel)+\sigma(IS+g_e\mu_B H_0)/2t \quad (28)$$

and

$$\Delta_\sigma=4\gamma(\mathbf{k}_\parallel)(1-t_s/t)-\sigma IS(1-I_s/I)/2t\ . \quad (29)$$

We note here that the second term of Eq. (27) represents the result for an *infinitely extended* system, whereas the first term is like a reflection term due to the presence of the surface. Equations (27)–(29) can now be substituted into (26) and, if the integrations over ω_1 and ω_2 are carried out, we have an expression for the spin-spin Green function $g^0_{mn}(\mathbf{k}_\parallel,\omega)$. From this, together with Eq. (24), we have a linear-response relation that can be used to relate the matrix Green function $\underline{G}'$ and $\underline{G}$ in the formalism of Sec. II and hence solve for the excitations of the system.

However, before proceeding with this calculation for a semi-infinite ferromagnetic semiconductor, it is helpful to examine briefly the corresponding results for an infinitely extended material. In this case the single-particle spectral weight for an electron with a three-dimensional wave vector $\mathbf{k}$ and spin projection σ can be simply written as the delta function $\frac{1}{2}\delta[\omega-W_e(\mathbf{k})+\sigma IS/2]$, with $W_e(\mathbf{k})$ denoting the band energy. When this is substituted into the three-dimensional analog of Eq. (26) we find

$$g^0(\mathbf{k},\omega)=\frac{1}{2\pi N'}\sum_{\mathbf{q}}\frac{f[W_e(\mathbf{q})-IS/2]-f[W_e(\mathbf{k}+\mathbf{q})+IS/2]}{\hbar\omega-IS-g_e\mu_B H_0-W_e(\mathbf{k}+\mathbf{q})+W_e(\mathbf{q})+i\varepsilon}\ , \quad (30)$$

where $\mathbf{q}$ is a three-dimensional wave vector and N' is the total number of spin sites. Using the linear response result in Eq. (24) and the Green-function equations of motion for the localized spin operators as in Sec. II (but applied now to an infinite system) we eventually obtain

$$\omega-g\mu_B H_0-I\langle s^z\rangle-2SJ[3-2\gamma(\mathbf{k}_\parallel)-\cos(k_z a)]-\pi SI^2 g^0(\mathbf{k},\omega)=0 \quad (31)$$

for a simple-cubic material, where $\mathbf{k}=(\mathbf{k}_\parallel,k_z)$. Hence this approach correctly reproduces the expression previously derived[21–23] (by other methods) for the excitation frequencies in an infinite bulk system. Equation (31) implies that the frequency of the lower spin wave at wave vector $\mathbf{k}=\mathbf{0}$ is simply $g\mu_B H_0$, which is independent of the interaction I as a consequence of the rotational symmetry of the Hamiltonian $\mathcal{H}_I$. Thus the approximations of our linear-response approach are consistent with this symmetry requirement.

In the present case of a semi-infinite system we note, by analogy with the discussion in Sec. II, that each of the Green-function components $g^0_{mn\sigma}(\mathbf{k}_\parallel,\omega)$ for the itinerant electrons provides a bulklike contribution for frequencies such that $|x_\sigma|=1$. Also, provided an existence condition is satisfied, there may be an additional surfacelike contribution when $|x_\sigma|<1$. In general, the corresponding spectral weights in Eq. (26) will contain both bulklike and surfacelike terms. The modified bulklike contribution to the spectral weight, obtained after formally rewriting x_σ in Eq. (27) as $\exp(ik_{z\sigma}a)$ with $k_{z\sigma}$ real, is

$$\rho^{0B}_{mn\sigma}(\mathbf{k}_\parallel,\omega)=[4\pi t\sin(k_{z\sigma}a)]^{-1}\times\left[\cos(k_{z\sigma}a|m-n|)-\frac{\cos[k_{z\sigma}a(m+n)]+2\Delta_\sigma\cos[k_{z\sigma}a(m+n-1)]+\Delta_\sigma^2\cos[k_{z\sigma}a(m+n-2)]}{1+2\Delta_\sigma\cos(k_{z\sigma}a)+\Delta_\sigma^2}\right], \quad (32)$$

where $k_{z\sigma}$ is related to the frequency ω through Eq. (28). The surfacelike contribution is obtained when x_σ is real and $|x_\sigma|<1$. The imaginary contribution to Eq. (27) in this case comes from the vanishing of its denominator for $x_\sigma=-\Delta_\sigma^{-1}$. The corresponding spectral weight (provided that the existence condition $|\Delta_\sigma|>1$ is satisfied) is

$$\rho^{0s}_{mn\sigma}(\mathbf{k}_\parallel,\omega)=-\frac{x_\sigma^{m+n}\Delta_\sigma^{-1}(1+x_\sigma^{-1}\Delta_\sigma)^2}{2(x_\sigma-x_\sigma^{-1})}\delta(\omega-\omega_\sigma^s)\ , \quad (33)$$

where we denote

$$\omega_\sigma^s=4t\gamma(\mathbf{k}_\parallel)-t(\Delta_\sigma+\Delta_\sigma^{-1})-\sigma(IS+g_e\mu_B H_0)/2\ . \quad (34)$$

We can now substitute the above expressions for the spectral weights into Eq. (26) to obtain $g^0_{mn}(\mathbf{k}_\parallel,\omega)$. We can finally use this along with Eq. (24) to write the matrix Green function $\underline{G}'(\mathbf{k}_\parallel,\omega)$ of Eq. (7) in terms of $\underline{G}(\mathbf{k}_\parallel,\omega)$ as

$$\underline{G}'(\mathbf{k}_\parallel,\omega)=-\pi I\underline{g}^0(\mathbf{k}_\parallel,\omega)\underline{R}\,\underline{G}(\mathbf{k}_\parallel,\omega)\ . \tag{35}$$

Substituting this in Eq. (7) we can solve for the magnetic Green function from the following formal expression:

$$\left[\underline{A}_0+\underline{D}_0+\pi\frac{I^2}{J}\underline{R}\,\underline{g}^0(\mathbf{k}_\parallel,\omega)\underline{R}\right]\underline{G}(\mathbf{k}_\parallel,\omega)=-\frac{1}{\pi J}\underline{I}_0\ . \tag{36}$$

The bulk and surface spin-wave frequencies correspond to the poles of $\underline{G}(\mathbf{k}_\parallel,\omega)$ from Eq. (36). For general values of the parameters t, t_s, I, and I_s, the matrix Green function $\underline{g}^0(\mathbf{k}_\parallel,\omega)$ is infinite dimensional and has all matrix elements nonzero. In this case Eq. (36) can always be solved numerically for $\underline{G}(\mathbf{k}_\parallel,\omega)$ by introducing a truncation approximation to reduce the dimension of the matrices to a finite value. However, in the next section we consider Eq. (36) in further detail for some particular choices of the model parameters and we show that an analytic solution is still possible in appropriate cases.

It should be pointed out here that the transverse spin susceptibility of Eq. (26) in the semi-infinite case has been derived before in the case of paramagnetic metals[26] (where $I=0$), and also as the zeroth-order Green function in the calculation of the Dyson series for ferromagnetic metals using the Hubbard model.[12,13] However, in all these calculations t_s was set equal to t, and hence the only effect of the surface was that an electron at the surface had fewer nearest neighbors compared with an electron in the bulk. Also Δ_σ was equal to zero, so there was no surfacelike contribution to the spectral weight as in Eq. (33) and the modified bulk-mode contribution of Eq. (32) was much simpler.

V. EXPANSIONS FOR SMALL t

We now obtain some analytical expressions for the case of the hopping parameter t being small. In order to further simplify the problem let us consider the case of $|\Delta_\sigma|<1$, where Δ_σ is defined in Eq. (29). If we assume $I_s=I$ for the present (see later), then the inequality is satisfied for all wave vectors $\mathbf{k}_\parallel$ provided $\frac{3}{4}<t_s/t<\frac{5}{4}$. This automatically rules out any surface-wave contribution to the spectral weight and we need to consider only the bulklike contribution. By substituting Eq. (32) into Eq. (26) and changing the integration variables from ω_1 and ω_2 to k_{z-} and k_{z+}, respectively, we obtain

$$g^0_{mn}(\mathbf{k}_\parallel,\omega)=\frac{a^2}{2\pi^3N}\sum_{\mathbf{q}_\parallel}\int_0^{\pi/a}dk_{z-}\int_0^{\pi/a}dk_{z+}\,C_{mn}(\mathbf{k}_{z-},\Delta_-)C_{nm}(k_{z+},\Delta_+)$$
$$\times\frac{f[W_e(\mathbf{q}_\parallel,k_{z+})-IS/2]-f[W_e(\mathbf{k}_\parallel+\mathbf{q}_\parallel,k_{z-})+IS/2]}{\hbar\omega-IS-g_e\mu_BH_0-W_e(\mathbf{k}_\parallel+\mathbf{q}_\parallel,k_{z-})+W_e(\mathbf{q}_\parallel,k_{z+})+i\varepsilon}\ , \tag{37}$$

where the band energies are given by

$$W_e(\mathbf{k}_\parallel,k_z)=2t[2\gamma(\mathbf{k}_\parallel a)+\cos(k_za)]\ , \tag{38}$$

and $C_{mn}(k_{z\sigma},\Delta_\sigma)$ are functions representing the combinations of cosine terms appearing within the curly brackets in Eq. (32). If we assume $t_s=t$ in addition to $I_s=I$, then Δ_σ vanishes and we have

$$C_{mn}(k_{z\sigma},\Delta_\sigma=0)=\cos(k_{z\sigma}a|m-n|)-\cos[k_{z\sigma}a(m+n)]\ . \tag{39}$$

If this is substituted into Eq. (37) and I is set equal to 0, we recover the previous result[26] established for a paramagnetic metal. Equation (37) with the full form of $C_{mn}(k_{z\sigma},\Delta_\sigma)$ for $\Delta_\sigma\neq0$ represents a generalization of this expression.

For small t such that $t\ll|\hbar\omega-IS-g_e\mu_BH_0|$ we can expand the denominator of Eq. (37) in a power series and carry out the integrations over k_{z-} and k_{z+} term by term. This leads to the following expansion, which is exact up to second order in t (or t_s):

$$g^0_{mn}(\mathbf{k}_\parallel,\omega)=\frac{\langle s^z\rangle}{\pi(\hbar\omega-IS-g_e\mu_BH_0)}[\delta_{m,n}+2t^2(\hbar\omega-IS-g_e\mu_BH_0)^{-2}(4\delta_{m,n}[1-\gamma(\mathbf{k}_\parallel)]\{1-[1-(t_s/t)^2]\delta_{m,l}\}$$
$$+\delta_{m,n}(2-\delta_{m,1})-\delta_{|m-n|,1})+O(t^4)]\ . \tag{40}$$

We notice from Eq. (40) that $g^0_{mn}(\mathbf{k}_\parallel,\omega)$ is tridiagonal (up to order t^2) and hence when it is incorporated into Eq. (36) we obtain the following formal matrix equation:

$$(\underline{A}_t+\underline{D}_t)\underline{G}(\mathbf{k}_\parallel,\omega)=-\frac{1}{\pi J(1+b)}\underline{I}_0\ , \tag{41}$$

where $\underline{A}_t$ and $\underline{D}_t$ are defined similarly to the matrices $\underline{A}_0$ and $\underline{D}_0$ in Sec. II, except that the quantities d_0 and δ_0 are replaced by

$$d_t=(1+b)^{-1}\{d_0+I^2\langle s^z\rangle/[J(\hbar\omega-IS-g_e\mu_BH_0)]+2b[3-2\gamma(\mathbf{k}_\parallel)]\}\ , \tag{42}$$

$$\delta_t=(1+b)^{-1}(\delta_0-b\{1+4[1-\gamma(\mathbf{k}_\parallel)][1-(t_s/t)^2]\})\ , \tag{43}$$

where

$$b=t^2[2I^2\langle s^z\rangle/J(\hbar\omega-IS-g_e\mu_BH_0)^3]\ . \tag{44}$$

We can now solve Eq. (41) by following the same matrix inversion method as described in Sec. III. The bulk spin-wave frequencies in the semi-infinite ferromagnetic semiconductor are obtained as the solutions of

$$\hbar\omega-g\mu_BH_0-2SJ[3-2\gamma(\mathbf{k}_\parallel)-\cos(k_za)]-I\langle s^z\rangle - \frac{I^2S\langle s^z\rangle}{\hbar\omega-IS-g_e\mu_BH_0}\left[1+\left[\frac{2t}{\hbar\omega-IS-g_e\mu_BH_0}\right]^2[3-2\gamma(\mathbf{k}_\parallel)-\cos(k_za)]\right]=0\ , \tag{45}$$

where k_z is a real wave-vector component perpendicular to the surface. The same result can, in fact, be deduced from a small t expansion of Eq. (31), for an *infinite* ferromagnetic semiconductor. The surface spin-wave frequencies satisfy the equation

$$\hbar\omega-g\mu_BH_0-2SJ[3-2\gamma(\mathbf{k}_\parallel)]-SJ(\delta_t+\delta_t^{-1})-I\langle s^z\rangle - \frac{I^2S\langle s^z\rangle}{\hbar\omega-IS-g_e\mu_BH_0}\left[1+\left[\frac{2t}{\hbar\omega-IS-g_e\mu_BH_0}\right]^2[3-2\gamma(\mathbf{k}_\parallel)+(\delta_t+\delta_t^{-1})/2]\right]=0\ , \tag{46}$$

provided the existence condition $|\delta_t|>1$ is satisfied. We notice immediately that when t and t_s are both set to zero in Eqs. (45) and (46) we recover the results of Sec. III for the case of $I_s=I$.

Some numerical examples to illustrate the results for $t\neq0$ are given in Fig. 3, where we plot $\hbar\omega/SJ$ against $\mathbf{k}_\parallel a$ for various combinations of the parameters J_s/J and t_s/t (taking $I_s/I=1$). The surface and bulk spin-wave frequencies are shown just for the low-frequency excitations, since the small t expansion is good in this regime. The effects of taking nonzero t and t_s values can be seen by comparing Fig. 3 with Fig. 1.

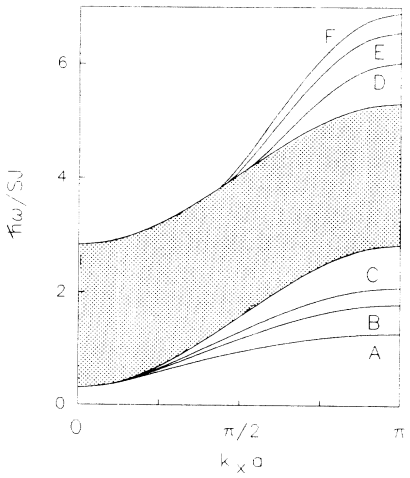

FIG. 3. The spin-wave frequencies (in units of $SJ/\hbar$) plotted against k_xa for a semi-infinite narrow-band semiconductor (assuming $t\neq0$) in the low-frequency region. The parameter values are $I/J=200$, $t/J=10$, $S=3/2$, $I_s/I=1$, $\langle s^z\rangle=\frac{1}{2}$, $g\mu_BH_0/SJ=0.3$, and $g_e=g$. The bulk spin-wave region is shaded, and the labeling of the surface spin-wave branches corresponds to *A*, $J_s/J=0.5$, $t_s/t=1.5$; *B*, $J_s/J=0.5$, $t_s/t=1$; *C*, $J_s/J=0.5$, $t_s/t=0.5$; *D*, $J_s/J=2$, $t_s/t=1.5$; *E*, $J_s/J=2$, $t_s/t=1$; *F*, $J_s/J=2$, $t_s/t=0.5$.

We now briefly discuss the case of a small t and $I\neq I_s$. In this case it can be seen from Eq. (29) that $|\Delta_\sigma|$ will be greater than unity, unless I and I_s are very close in value. Hence, in general both the bulk and surface parts of the spectral weights [see Eqs. (32) and (33)] have to be taken into account in the calculation of $\underline{g}^0(\mathbf{k}_\parallel,\omega)$. There will be four terms to evaluate in Eq. (26), one involving the bulklike parts of the spectral weights, one involving the surfacelike parts, and two cross terms. However, there is a simplification in the limit when t and t_s vanish, giving $|\Delta_\sigma|\to\infty$. It may be verified that the cross terms vanish and Eq. (26) reduces to

$$g^0_{mn}(\mathbf{k}_\parallel,\omega)=\frac{\langle s^z\rangle\delta_{m,1}\delta_{n,1}}{\pi(\hbar\omega-I_sS-g_e\mu_BH_0)} + \frac{\langle s^z\rangle(\delta_{m,n}-\delta_{m,1}\delta_{n,1})}{\pi(\hbar\omega-IS-g_e\mu_BH_0)}\ , \tag{47}$$

where the first and second terms arise from the surfacelike and bulklike parts of the spectral weights, respectively. When Eq. (47), which is valid as $t\to0$, is substituted into Eq. (36) we correctly recover the results of Sec. III. Thus we conclude that the methods described in Sec. IV are quite general and are applicable for both wide-band ($W\gg IS$) and narrow-band ($W\ll IS$) semiconductors.

VI. DISCUSSION AND CONCLUSIONS

In this paper we have concentrated on evaluating the spin-wave excitations. However, the poles of the two-particle Green functions $\underline{g}^0(\mathbf{k}_\parallel,\omega)$ provide the collective

modes in the system. In an infinite bulk system they give rise to the so-called Stoner continuum which is a superposition of electron-hole excitations of opposite spin. We now indicate briefly how these excitations would be modified in the presence of a surface. It is apparent from Eq. (26) that the poles of $\underline{g}^0(\mathbf{k}_\parallel,\omega)$ occur at $\omega=\Omega(\mathbf{k}_\parallel,\mathbf{q}_\parallel)$, where

$$\Omega(\mathbf{k}_\parallel,\mathbf{q}_\parallel)=\omega^{B,S}_{\sigma=-1}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)-\omega^{B,S}_{\sigma=1}(\mathbf{q}_\parallel)\ . \tag{48}$$

The superscripts B and S refer to the bulklike and surfacelike contributions to the one-particle excitation frequencies, respectively. We have

$$\omega^B_\sigma(\mathbf{k}_\parallel)=2t\,[2\gamma(\mathbf{k}_\parallel)+\cos(k_z a)]-\sigma(IS+g_e\mu_B H_0)/2\ , \tag{49}$$

and ω^S_σ is given by Eq. (34). All the various combinations of bulk- and surface-type terms in Eq. (48) are possible, provided the existence condition $|\Delta_\sigma|>1$ for a surface excitation is satisfied. A more detailed investigation of the spectrum of $\Omega(\mathbf{k}_\parallel,\mathbf{q}_\parallel)$ would be relevant if we wished to study the lifetimes of the spin-wave excitations, because a possible decay mechanism (as in *infinite* ferromagnetic semiconductors) for a spin wave would be for it to decay into a Stoner-type excitation of the same frequency.

We have obtained the dispersion relations for the spin waves in a semi-infinite ferromagnetic semiconductor described by the s-d (or s-f) interaction model, extending earlier work on infinite ferromagnetic semiconductors. The results in the case of narrow-band materials, such as the chromium spinels, have been emphasized. Several extensions of the present work are possible. These include further work on wide-band materials, for which we already have formal results in Sec. IV, to obtained detailed numerical applications to specific materials such as EuO or EuS. Other extensions of this work include the study of surface excitations in antiferromagnetic semiconductors and in certain diluted magnetic semiconductors.

ACKNOWLEDGMENTS

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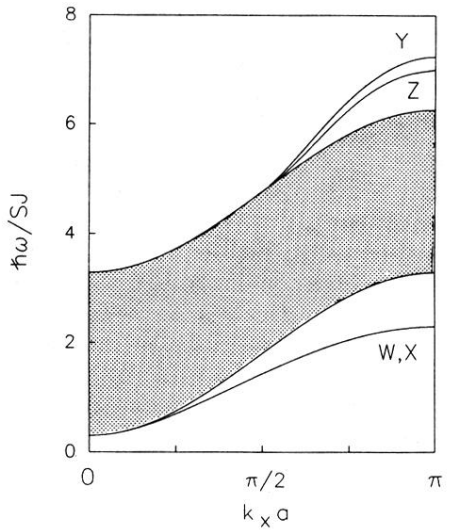

FIG. 1. The spin-wave frequencies (in units of $SJ/\hbar$) plotted against $k_x a$ for a semi-infinite narrow-band semiconductor (with $t=0$) in the low-frequency region. The parameter values are $I/J=200$, $S=\frac{3}{2}$, $\langle s^z\rangle=\frac{1}{2}$, $g\mu_B H_0/SJ=0.3$, and $g_e=g$. The bulk spin-wave region is shaded, and the labeling of the surface spin-wave branches corresponds to W, $J_s/J=0.5$, $I_s/I=1$; X, $J_s/J=0.5$, $I_s/I=0.2$; Y, $J_s/J=2$, $I_s/I=1$; Z, $J_s/J=2$, $I_s/I=0.2$.

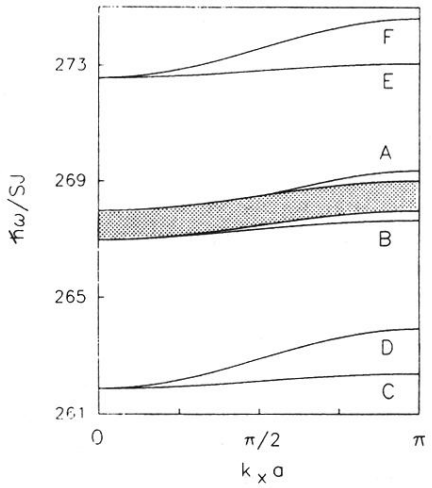

FIG. 2. As in Fig. 1, but in the high-frequency region. In this case the labeling of the surface spin-wave branches corresponds to *A*, $J_s/J=2$, $I_s/I=1$; *B*, $J_s/J=0.5$, $I_s/I=1$; *C*, $J_s/J=0.5$, $I_s/I=0.98$; *D*, $J_s/J=2$, $I_s/I=0.98$; *E*, $J_s/J=0.5$, $I_s/I=1.02$; *F*, $J_s/J=2$, $I_s/I=1.02$.

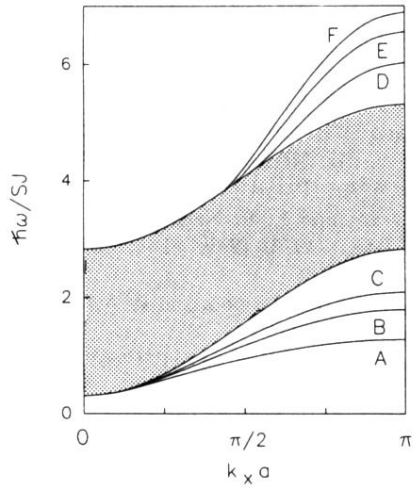

FIG. 3. The spin-wave frequencies (in units of $SJ/\hbar$) plotted against $k_x a$ for a semi-infinite narrow-band semiconductor (assuming $t \neq 0$) in the low-frequency region. The parameter values are $I/J = 200$, $t/J = 10$, $S = 3/2$, $I_s/I = 1$, $\langle s^z \rangle = \frac{1}{2}$, $g\mu_B H_0/SJ = 0.3$, and $g_e = g$. The bulk spin-wave region is shaded, and the labeling of the surface spin-wave branches corresponds to *A*, $J_s/J = 0.5$, $t_s/t = 1.5$; *B*, $J_s/J = 0.5$, $t_s/t = 1$; *C*, $J_s/J = 0.5$, $t_s/t = 0.5$; *D*, $J_s/J = 2$, $t_s/t = 1.5$; *E*, $J_s/J = 2$, $t_s/t = 1$; *F*, $J_s/J = 2$, $t_s/t = 0.5$.