Inelastic scattering of low-energy electrons by spin excitations on ferromagnets

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We develop a multiple-scattering description of the inelastic scattering by spin excitations of low-energy electrons reflected from the surface of a ferromagnetic material. The scattering efficiency per unit solid angle is expressed in terms of an appropriately defined frequency- and wave-vector-dependent susceptibility of the near surface region of the crystal. The formalism may thus be applied to the scattering by either Stoner excitations, or spin waves, provided the appropriate dynamic response functions are available. We report quantitative studies of the excitation cross sections for spin waves on the Fe(100) surface, and on the Ni(110) surface. The scattering efficiencies we calculate are compared to those for exciting (short-wavelength) surface phonons on such surfaces, in electron-loss studies in the impact regime. We then comment on possible application of electron-loss spectroscopy to the study of spin excitations on magnetic surfaces, and in ultrathin films.

I. INTRODUCTION

Low-energy electrons with energies in the range of a few to a few hundred electron volts are a powerful means of probing the outermost atomic layers of crystals and ultrathin films adsorbed on surfaces. Their surface sensitivity has its origin in the short mean free path in matter in this energy range, which is typically two or three interatomic spacings. During the past decade , electron-energy-loss spectroscopy[1] has been employed to explore the dispersion relations of diverse elementary excitations localized near crystal surfaces, such as surface phonons[2] and surface plasmons.[3]

Considerably less attention has been devoted to the use of electron-energy-loss spectroscopy to explore the nature of magnetic excitations on ferromagnets, or in ultrathin magnetically ordered films on substrates. While various spin-polarized electron spectroscopies have developed impressively during the past few years, and, in fact, spin-polarized electron-energy-loss spectroscopy has been employed to examine losses produced by spin-flip excitations in ferromagnets,[4] rather little theoretical activity has been directed toward this area.

The purpose of this paper is to develop a general description of the energy loss suffered by electrons reflected from a magnetic surface, by processes which flip the spin of the beam electron. This is done within the framework of a multiple-scattering description of the electron-substrate interaction. One may view this analysis as the extension to the scattering of incoming electrons by magnetic degrees of freedom of an earlier formalism developed to describe the excitation of phonons on the crystal surface by low-energy electrons.[5,6] By now, the phonon cross-section analyses have been applied with very considerable success to the quantitative analysis of electron-loss data, in the off specular geometry, for electrons incident on rather diverse substrates.[7–11] The success realized by electron-loss spectroscopy in the study of the dispersion relations of surface phonons is due, in part, to theory and its ability to provide quantitative interpretation of the loss spectra. It is our hope that the development and implementation of calculations such as those reported here will prove as useful in the study of the magnetic degrees of freedom at surfaces and in ultrathin films.

There are distinct differences between the questions that need to be addressed by theory in the area of surface and thin-film magnetism, compared to those which arise in surface lattice dynamics. In the latter case, at least at temperatures that are modest, lattice dynamics in the harmonic approximation suffices to describe the frequency spectrum of atomic motions in and near the surface. The analysis of the electron-energy-loss spectrum is cast conveniently in terms of the eigenvectors of the various surface and bulk phonons. In the magnetic systems, most particularly itinerant electron magnets such as the $3d$ transition metals, the excitation spectrum is more complex and less well understood. While very general considerations suggest that at long wavelengths the magnetic excitations are spin waves in such systems,[12] at short wavelengths (in the bulk) we encounter Stoner excitations (particle-hole excitations in which the spin of the excited electron flips) in the response function, whose spectral shape is influenced by electron-electron interactions. The spin-wave dispersion curve may overlap the Stoner continuum, with the consequence that the short-wavelength modes suffer appreciable Landau damping.[13] The nature of the elementary excitations at the surface or in an ultrathin film of such a material has yet to be explored theoretically; as remarked earlier, electron-loss studies to date report Stoner excitations but reveal no spin waves.

Considerations such as those given above lead us to present a discussion of the inelastic scattering in general terms, without resort to a particular model of spin excitations at the surface. We arrive at an expression for the loss cross section that is expressed in terms of certain wave vector and frequency-dependent response functions of the outermost atomic layers of the material. We envision its application to theoretical descriptions of magnetic excitations near the surface of itinerant electron

materials, but in this paper we confine our attention to the case where the elementary excitations are supposed to have the character of spin waves appropriate to a localized spin model. We report calculations of the magnitude and energy dependence of the spin-wave excitation cross section, for such spin waves on the Fe(100) and Ni(110) surface. The results, when compared to earlier calculations of the surface phonon excitation cross sections, provide us with quantitative estimates of the relative magnitude of the absolute cross section for losses from magnetic excitations. With these reults in hand, we discuss future prospects for more detailed experimental studies of magnetic excitations in ultrathin films, and at magnetic surfaces.

Our analysis assumes the beam electron senses the thermal fluctuations in orientation of the substrate magnetic moment through the exchange interaction between the incoming electron, and the "magnetically active" electrons in the substrate. More precisely, we model the substrate within a muffin-tin picture. Inside each muffin tin, we have a spin-dependent potential one may write $V(r)I+B(r)\hat{n}$, with $\hat{n}$ the instantaneous direction of the local moment. Fu and Freeman have kindly provided us with explicit forms for $V(r)$ and $B(r)$, from their *ab initio* studies of Fe and Ni films. It should be noted that, in previous papers, we have found that, for the Fe(110) surface, these potentials provide a remarkably quantitative account of spin-polarized low-energy electron-diffraction (SPLEED) data over a wide range of beam energies,[14] with no need to introduce modifications of these ground-state potentials. These eariler calculations also suppose the beam electron mean free path in the substrate has a negligible dependence on spin orientation, an assumption reinforced by our theoretical interpretation of the spin dependence of photoelectron transmission through ultrathin films of Fe on Cu(100).[15] The success of these earlier studies provides us with some confidence that the excitation cross-section calculations reported here are indeed reliable from the quantitative point of view.

II. THEORETICAL FORMALISM

A. General discussion

In the present section we consider the inelastic scattering of spin-polarized electrons off the spin excitations of a two-dimensionally periodic ferromagnetic crystal. In the model we consider, the multiple-scattering aspects of the problem are treated within the range of validity of the muffin-tin picture and the adiabatic approximation, wherein the motions of the spins in the crystal are regarded as slow. We shall focus on obtaining a general expression for the scattering efficiency per unit solid angle per loss energy for the case in which a *single* inelastic spin-flip event is experienced by the beam electron within the substrate. Later we show how this description, phrased in terms of certain wave-vector- and frequency-dependent susceptibilities of the surface region, may be applied to the calculation of the cross section for exciting spin waves. It is to be noted that aspects of the following derivation closely parallel that of the scattering efficiency for electron energy loss due to phonons [electron-energy-loss spectroscopy (EELS)] and earlier low-energy electron diffraction (LEED) work,[5,16] so that, in the present analysis, we focus primarily on the new aspects of the problem.

We begin by considering the structure of the electron-substrate scattering amplitude. The state vector of the electron-substrate system is expressed as a product of the beam electron state vector and substrate state vector, so that, if we let $|i\rangle$ and $|s\rangle$ ($|I\rangle$ and $|F\rangle$) denote the initial and final states of the incident electron (substrate), then we can express the scattering amplitude M by the formal expression

$$M=\langle F|\langle s|GT|i\rangle|I\rangle\equiv\langle F|f|I\rangle\ , \tag{1}$$

where G denotes the single-particle propagator and T denotes the scattering "T-matrix" for the entire crystal. The "T matrix" is, of course, defined through the usual relation[16]

$$V|\Psi\rangle=T|i\rangle|I\rangle\ , \tag{2}$$

where $|\Psi\rangle$ denotes the exact electron-substrate state vector and V denotes the potential-energy operator for the electron-substrate system. For the magnetic problem, we assume the electron interacts with each substrate atom via a spin-dependent interaction that contains exchange so that, in the present muffin-tin picture, V is an operator in the spin space of the electron having the form

$$V=\sum_{\mathbf{R}} v_{\mathbf{R}(l)}[|\mathbf{r}-\mathbf{R}(l)|;\boldsymbol{\sigma}\cdot\mathbf{S}(l)]\ , \tag{3}$$

where $\boldsymbol{\sigma}$ is the spin of the incident electron, $\mathbf{R}(l)$ is the coordinate vector for the atom at site l, $\mathbf{r}$ is the incident electron's coordinate vector, and $\mathbf{S}(l)$ is the spin moment at site l. There are two assumptions built into Eq. (3). The first, tested and found to work very well in our earlier calculations of elastic scattering from Fe(110),[14,15] is that the potential may be taken to be spherically symmetric. The second is an adiabatic approximation. As a thermal fluctuation reorients a particular moment, from, say, the $\hat{z}$ direction along which it is aligned in the ground state to the direction $\mathbf{S}(l)$, the spin-dependent portion of the potential just follows the spin. We shall assume it rotates rigidly.

Now, magnetic excitations produce "small" changes in the spin moment vector $\mathbf{S}(l)$ so that the scattering amplitude may be expanded formally as

$$M=\langle F|f|I\rangle$$
$$=\langle F|f_0|I\rangle+\sum_{\alpha,l}\left[\frac{\partial f}{\partial S_\alpha(l)}\right]_0\langle F|\delta S_\alpha(l)|I\rangle+\cdots\ . \tag{4}$$

In this expression, the subscript zero denotes evaluation at the equilibrium state of the crystal and we can take $\alpha=+$ or $-$ where the "ladder" operators $S_\pm(l)$ are given by the usual expression $S_\pm=S_x\pm iS_y$. The first term is the amplitude calculated in SPLEED theory. For the present process, this is indentically zero because $|F\rangle$ describes a state of the substrate in which a spin excitation has been created. We only consider the term linear

in $\delta S_\alpha(l)$ since only it gives rise to *single* excitation processes. If $|I\rangle$ is the ferromagnetic ground state with moments aligned along $+\hat{z}$, the term $\langle F|\delta S_+(l_\parallel,l_z)|I\rangle$ vanishes. At finite temperatures, the beam electron may encounter the substrate in an excited state rather than the magnetic ground state, and a detailed balancing argument relates the cross section from this term to that generated by $\langle F|\delta S_-(l_\parallel,l_z)|I\rangle$. Thus, in what follows, we consider only losses generated by $\delta S_-(l_\parallel,l_z)$.

We now describe the geometry to be considered. In Fig. 1 we show that the crystal can be supposed to be constructed of unit cells semi-infinite in length normal to the surface. We label the two-dimensional translational vector between the unit cell at $l_\parallel$ and a reference cell 0 as $\mathbf{R}_\parallel(l_\parallel)$. Clearly, this vector lies in a plane parallel to the surface. Each unit cell consists of atoms within the layers that are parallel to the surface under study. We use l_z to describe the location of one of these layers. Additionally, it is assumed that each layer consists of atoms having the same spin moment and that each layer contributes one atom only to the semi-infinite unit cell. The general case of two or more distinct coplanar atoms can be treated by a "spin-polarized" version of the combined space method.[17]

Thus, with this geometry, if we write l as $l_\parallel+l_z\hat{z}$ (where $\hat{z}$ is in the direction normal to the surface) and use the expansion

$$\delta S_-(l_\parallel,l_z)=\sum_{\mathbf{q}_\parallel}\frac{1}{\sqrt{N_s}}\exp[-i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel(l_\parallel)]S_-(\mathbf{q}_\parallel,l_z)\ , \tag{5}$$

where N_s denotes the number of unit cells in the basic quantization area, then we can express the scattering amplitude for a single inelastic event associated with wave-vector transfer $\mathbf{q}_\parallel$ as

$$M(\mathbf{q}_\parallel)=\sum_{l_\parallel,l_z}\frac{1}{\sqrt{N_s}}\exp[-i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel(l_\parallel)]\left[\frac{\partial f}{\partial S_-(l)}\right]_0\times\langle F|S_-(\mathbf{q}_\parallel,l_z)|I\rangle\ . \tag{6}$$

Now we must consider the structure of $(\partial f/\partial S_-(l))_0$,

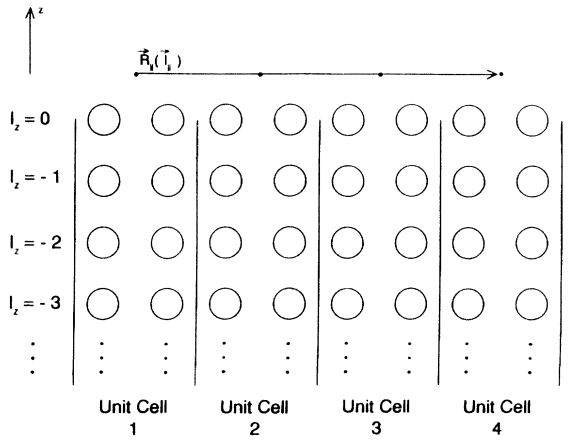

FIG. 1. An illustration of the geometry which forms the basis of the present analysis. We regard the crystal as a two-dimensionally periodic array of unit cells, each semi-infinite in length. Here we depict two atoms per layer per unit cell.

within the framework of multiple-scattering theory. Since $f=\langle s|GT|i\rangle$, we have

$$\left[\frac{\partial f}{\partial S_-(l)}\right]_0=\langle s|G\left[\frac{\partial T}{\partial S_-(l)}\right]_0|i\rangle\ . \tag{7}$$

In the spirit of EELS,[5] we define a spin-polarized EELS wave function for a particular layer l_z through

$$\langle\mathbf{r}\sigma|\Psi_{\rm SP}^{(l_z)}\rangle=\frac{1}{\sqrt{V}}\sum_{l_\parallel}\exp[-i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel(l_\parallel)]\times\left\langle\mathbf{r}\sigma\left|G\left[\frac{\partial T}{\partial S_-(l)}\right]_0\right|\mathbf{k}_i\sigma_i\right\rangle\ , \tag{8}$$

where, to introduce a more explicit notation, we have let $|i\rangle=|\mathbf{k}_i\sigma_i\rangle$. The incident beam electron thus has vector $\mathbf{k}_i$ and spin σ_i. In coordinate and spin space, this becomes[5]

$$\langle\mathbf{r}\sigma|\Psi_{\rm SP}^{(l_z)}\rangle=\frac{1}{\sqrt{V}}\sum_{l_\parallel}\exp[-i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel(l_\parallel)]\frac{2m}{\hbar^2}\times\int\frac{d^3k}{(2\pi)^3}\frac{\exp(i\mathbf{k}\cdot\mathbf{r})}{D^\sigma(k,E)}\int d^3r_1d^3r_2\exp(-i\mathbf{k}\cdot\mathbf{r}_1)\left[\frac{\partial T(\mathbf{r}_1\mathbf{r}_2;\{\mathbf{R}(l)\},\{\mathbf{S}(l)\})}{\partial S_-(l)}\right]_0^{\sigma\sigma_i}\exp(i\mathbf{k}_i\cdot\mathbf{r}_2)\ , \tag{9}$$

where we have used $\{\mathbf{R}(l)\}$ and $\{\mathbf{S}(l)\}$ to denote dependence on the collection of all lattice and spin moment vectors, and

$$D^\sigma(k,E)=\frac{2m}{\hbar^2}[E-\Sigma^\sigma(k,E)]-k^2$$

with E and Σ^σ being the incident energy and the self-energy of the electron, respectively. The imaginary part of the self-energy leads to a finite mean free path for the beam electron. For convenience we write the scattered electron wave function as

$$\langle\mathbf{r}\sigma|\Psi_{\rm SP}^{(l_z)}\rangle=\frac{1}{\sqrt{V}}\frac{2m}{\hbar^2}\int\frac{d^3k}{(2\pi)^3}\frac{\exp(i\mathbf{k}\cdot\mathbf{r})}{D^\sigma(k,E)}\sum_{l_\parallel}\exp[-i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel(l_\parallel)]A_l(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)\ , \tag{10}$$

where

$$A_l(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)=\int d^3r_1 d^3r_2 \exp(-i\mathbf{k}\cdot\mathbf{r}_1)\left[\frac{\partial T(\mathbf{r}_1\mathbf{r}_2;\{\mathbf{R}(l)\},\{\mathbf{S}(l)\})}{\partial S_-(l)}\right]_0^{\sigma\sigma_i}\exp(i\mathbf{k}_i\cdot\mathbf{r}_2) \tag{11}$$

is the matrix element that links the incident plane wave to the outgoing scattered wave.

We can now obtain an important identity for $A_l(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)$ by employing the two-dimensional periodicity of the slab geometry. In the expression above, if we let $\mathbf{r}_i\rightarrow\mathbf{r}_i+\mathbf{R}_\parallel(l_\parallel)$, where $\mathbf{R}_\parallel(l_\parallel)$ is the vector described earlier, then, since

$$\left[\frac{\partial T(\mathbf{r}_1\mathbf{r}_2;\{\mathbf{R}(l)\},\{\mathbf{S}(l)\})}{\partial S_-(l)}\right]_0^{\sigma\sigma_i}$$

must be invariant under translation of *all* arguments by a planar lattice vector, it is evident that

$$\begin{aligned}A_l(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)&=\exp[-i(\mathbf{k}-\mathbf{k}_i)\cdot\mathbf{R}_\parallel(l_\parallel)]A_{l_\parallel=0,l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)\\&\equiv\exp[-i(\mathbf{k}-\mathbf{k}_i)\cdot\mathbf{R}_\parallel(l_\parallel)]A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)\ .\end{aligned} \tag{12}$$

Thus, we can write $\langle\mathbf{r}\sigma|\Psi_{\rm SP}^{(l_z)}\rangle$ as

$$\langle\mathbf{r}\sigma|\Psi_{\rm SP}^{(l_z)}\rangle=\frac{1}{\sqrt{V}}\frac{2m}{\hbar^2}\int\frac{d^3k}{(2\pi)^3}\frac{\exp(i\mathbf{k}\cdot\mathbf{r})}{D^\sigma(k,E)}\sum_{l_\parallel}\exp[-i(\mathbf{k}-\mathbf{k}_i+\mathbf{q}_\parallel)\cdot\mathbf{R}_\parallel(l_\parallel)]A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)\ . \tag{13}$$

Further, since the sum on $l_\parallel$ yields the parallel momentum-conserving δ function, we obtain a result that may be expressed as

$$\langle\mathbf{r}\sigma|\Psi_{\rm SP}^{(l_z)}\rangle=\frac{1}{\sqrt{V}}\frac{2m}{\hbar^2}\int\frac{d^3k}{(2\pi)^3}\frac{\exp(i\mathbf{k}\cdot\mathbf{r})}{D^\sigma(k,E)}\sum_{\mathbf{g}}\frac{(2\pi)^2}{A_c}\delta(\mathbf{k}_\parallel-\mathbf{k}_{i\parallel}+\mathbf{q}_\parallel+\mathbf{g})A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)\ , \tag{14}$$

where A_c is the area of a unit cell and $\mathbf{g}$ denotes a reciprocal-lattice vector.

Now we must obtain an explicit representation of $A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)$. It is shown in Appendix A that

$$\begin{aligned}A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)=-(4\pi)^2\sum_{LL'}Y_L(\hat{k})\sum_{z_1z_2}\{&[1-\tau(\mathbf{k})G(\mathbf{k})]^{-1}_{z_1z}\exp(-i\mathbf{k}\cdot\mathbf{R}_z)\\&\times Q(l_z)\exp(i\mathbf{k}_i\cdot\mathbf{R}_z)[1-G(\mathbf{k}_i)\tau(\mathbf{k}_i)]^{-1}_{zz_2}\}_{L\sigma,L'\sigma_i}Y_L^*(\hat{k}_i)\ ,\end{aligned} \tag{15}$$

where

$$Q(l_z)=(1-t_zG^{zz})^{-1}I_{l_z}(1-G^{zz}t_z)^{-1}$$

with t_z being the t matrix associated with layer l_z, G and τ are as defined in Appendix A, and I_{l_z} is the loss integral, which we evaluate in Sec. II B. Thus, with this expression, we obtain the following for $\langle\mathbf{r}\sigma|\Psi_{\rm SP}^{(l_z)}\rangle$:

$$\begin{aligned}\langle\mathbf{r}\sigma|\Psi_{\rm SP}^{(l_z)}\rangle=\frac{1}{\sqrt{V}}\frac{2m}{\hbar^2}\int\frac{d^3k}{(2\pi)^3}\frac{\exp(i\mathbf{k}\cdot\mathbf{r})}{D^\sigma(k,E)}\sum_{\mathbf{g}}\frac{(2\pi)^2}{A_c}\delta(\mathbf{k}_\parallel-\mathbf{k}_{i\parallel}+\mathbf{q}_\parallel+\mathbf{g})[-(4\pi)^2]\\\times\sum_{LL'}Y_L(\hat{k})\sum_{z_1z_2}\{[1-\tau(\mathbf{k})G(\mathbf{k})]^{-1}_{z_1z}\exp(-i\mathbf{k}\cdot\mathbf{R}_z)Q(l_z)\\\times\exp(i\mathbf{k}_i\cdot\mathbf{R}_z)[1-G(\mathbf{k}_i)\tau(\mathbf{k}_i)]^{-1}_{z_1z_2}\}_{L\sigma,L'\sigma_i}Y_L^*(\mathbf{k}_i)\ .\end{aligned} \tag{16}$$

The integral on $\mathbf{k}_\parallel$ can be done trivially, however, the integral on $k_\perp$ requires a little work. The integral is done in the complex $k_\perp$ plane. The calculation is fairly straightforward and has been done elsewhere;[16] the result is

$$\begin{aligned}\langle\mathbf{r}\sigma|\Psi_{\rm SP}^{(l_z)}\rangle=-\frac{1}{\sqrt{V}}\gamma_0^\sigma\exp[-i\mathbf{k}^-(\mathbf{g})\cdot\mathbf{r}]\sum_{\mathbf{g}}\sum_{LL'}\frac{Y_L[\mathbf{k}^-(\mathbf{g})]}{|k_\perp(\mathbf{g})|}\sum_{z_1z_2}(\{1-\tau[\mathbf{k}^-(\mathbf{g})]G[\mathbf{k}^-(\mathbf{g})]\}^{-1}_{z_1z}\exp[-i\mathbf{k}^-(\mathbf{g})\cdot\mathbf{R}_z]Q(l_z)\\\times\exp(i\mathbf{k}_i\cdot\mathbf{R}_z)[1-G(\mathbf{k}_i)\tau(\mathbf{k}_i)]^{-1}_{zz_2})_{L\sigma,L'\sigma_i}Y_L^*(\hat{k}_i)\ ,\end{aligned} \tag{17}$$

where $\gamma_0^\sigma=-(8\pi^2i/A_c)(2m/\hbar^2)F^\sigma(k_0)$ (the function F^σ is defined in Appendix A), $k_0=\sqrt{(2m/\hbar^2)[E-\Sigma^\sigma(k_i,E)]}$, and $\mathbf{k}^\pm(\mathbf{g})=[\mathbf{k}_{i\parallel}-\mathbf{q}_\parallel+\mathbf{g},\pm\hat{z}k_\perp(\mathbf{g})]$ with $k_\perp(\mathbf{g})=\sqrt{k_0^2-k_\parallel^2(\mathbf{g})}$.

Now, the scattering amplitude is given by

$$M(\mathbf{q}_\parallel)=\sum_{lz}\frac{1}{\sqrt{N_s}}\int d^3r\langle \mathbf{k}_f\sigma_f|\mathbf{r}\sigma_f\rangle\langle \mathbf{r}\sigma_f|\Psi_{\mathrm{SP}}^{(l_z)}\rangle\langle F|S_-(\mathbf{q}_\parallel,l_z)|I\rangle \ . \tag{18}$$

The integral merely gives the condition that $\mathbf{k}_f=\mathbf{k}_f^{\pm}(\mathbf{g})=[\mathbf{k}_{i\parallel}-\mathbf{q}_\parallel+\mathbf{g},\pm\hat{z}k_{f\perp}(\mathbf{g})]$. Thus, the scattering amplitude is given by

$$M(\mathbf{q}_\parallel)=-\gamma_0^\sigma\frac{1}{\sqrt{N_s}}\sum_{lz}\sum_{\mathbf{g}}\sum_{LL'}\frac{Y_L[\mathbf{k}_f^-(\mathbf{g})]}{|k_{f\perp}(\mathbf{g})|}\sum_{z_1z_2}(\{1-\tau[\mathbf{k}_f^-(\mathbf{g})]G[\mathbf{k}_f^-(\mathbf{g})]\}_{z_1z}^{-1}\exp[-i\mathbf{k}_f^-(\mathbf{g})\cdot\mathbf{R}_z]Q(l_z)$$
$$\times\exp(i\mathbf{k}_i\cdot\mathbf{R}_z)[1-G(\mathbf{k}_i)\tau(\mathbf{k}_i)]_{zz_2}^{-1})_{L\sigma_f,L'\sigma_i}Y_L^*(\mathbf{k}_i)\langle F|S_-(\mathbf{q}_\parallel,l_z)|I\rangle \ . \tag{19}$$

We now want to use this expression for the scattering amplitude to develop an expression for the scattering efficiency per unit solid angle per unit loss energy. We denote the fractional number of electrons which emerge onto the solid angle $d\Omega$ after being inelastically scattered off a magnetic excitation of mode $(\mathbf{q}_\parallel)$ in the energy range $d\omega(d^2P/d\Omega\,d\omega)$. We can obtain this quantity by multiplying $|M|^2$ by the ratio of final to initial electron beam flux, summing over $\mathbf{k}_{f\parallel}$ in $d\Omega$, multiplying by the energy-conserving δ function $\delta[E_f-E_i+\hbar\omega(\mathbf{q}_\parallel)]$ and the initial substrate ensemble probabilities P_I, and then summing the result overall initial and final states of the substrate. The result is

$$\left[\frac{d^2P}{d\Omega\,d\omega}\right]=\left[\frac{mE_i\cos^2\theta_f}{2\pi^2\hbar^2\cos\theta_i}\right]A_c\sum_{l_zl_z'}\sum_{gg'}A_{l_z}^{(g)}(\mathbf{k}_f\sigma_f,\mathbf{k}_i\sigma_i)\delta[E_f-E_i+\hbar\omega(\mathbf{q}_\parallel)]A_{l_z'}^{(g')*}(\mathbf{k}_f\sigma_f,\mathbf{k}_i\sigma_i)$$
$$\times\sum_{I,F}\langle I|S_+(\mathbf{q}_\parallel,l_z')|F\rangle\langle F|S_-(\mathbf{q}_\parallel,l_z)|I\rangle P_I \ , \tag{20}$$

where θ_i and θ_f are the angles the initial and final electron beam make relative to the surface normal, A_c is the surface area of the sample, and $A_{l_z}^{(g)}(\mathbf{k}_f\sigma_f,\mathbf{k}_i\sigma_i)$ is defined as

$$A_{l_z}^{(g)}(\mathbf{k}_f\sigma_f,\mathbf{k}_i\sigma_i)=-\gamma_0^\sigma\sum_{LL'}\frac{Y_L[\hat{k}_f^-(\mathbf{g})]}{|k_{f\perp}(\mathbf{g})|}\sum_{z_1z_2}(\{1-\tau[\mathbf{k}_f^-(\mathbf{g})]G[k_f^-(\mathbf{g})]\}_{z_1z}^{-1}\exp[-ik_f^-(\mathbf{g})\cdot\mathbf{R}_z]Q(l_z)$$
$$\times\exp(i\mathbf{k}_i\cdot\mathbf{R}_z)[1-G(\mathbf{k}_i)\tau(\mathbf{k}_i)]_{zz_2}^{-1})_{L\sigma_f,L'\sigma_i}Y_L^*(\hat{k}_i) \ . \tag{21}$$

If we write

$$\delta[E_f-E_i+\hbar\omega(\mathbf{q}_\parallel)]=\int_{-\infty}^{\infty}\frac{dt}{2\pi\hbar}\exp\{i[\omega_{if}+\omega(\mathbf{q}_\parallel)]t\} \ ,$$

where $\omega_{if}=(E_f-E_i)/\hbar$, then we can express $(d^2P/d\Omega\,d\omega)$ in the final form

$$\left[\frac{d^2P}{d\Omega\,d\omega}\right]=\left[\frac{mE_i\cos^2\theta_f}{2\pi^2\hbar^2\cos\theta_i}\right]A_c\sum_{lzlz'}\sum_{gg'}A_{l_z}^{(g)}(\mathbf{k}_f\sigma_f,\mathbf{k}_i\sigma_i)A_{l_z'}^{(g')*}(\mathbf{k}_f\sigma_f,\mathbf{k}_i\sigma_i)$$
$$\times\int_{-\infty}^{\infty}\frac{dt}{2\pi\hbar}\exp(i\omega_{if}t)\langle S_+(\mathbf{q}_\parallel,l_z',t)S_-(\mathbf{q}_\parallel,l_z,0)\rangle \ , \tag{22}$$

where $S_+(\mathbf{q}_\parallel,l_z',t)$ is the spin moment operator in the Heisenberg representation and the quantity $\langle S_+(\mathbf{q}_\parallel,l_z',t)S_-(\mathbf{q}_\parallel,l_z,0)\rangle$ denotes the autocorrelation function of the spin moment operator. The angular brackets denote a statistical average over the appropriate finite-temperature statistical ensemble. This concludes our derivation of the scattering effficiency per solid angle per loss energy.

As mentioned in Sec. I, we have arrived at an expression for the scattering effficiency, without resort to any particular physical picture of the spin dynamics of the substrate. Thus, the expression can be applied to itinerant electron systems, and the Stoner excitations therein, or to a localized spin model of the Heisenberg type. Within any such picture, one's task is to evaluate the spin autocorrelation function $\langle S_+(\mathbf{q}_\parallel,l_z',t)S_-(\mathbf{q}_\parallel,l_z,0)\rangle$ displayed in Eq. (22). Later in this paper we shall do this for the Heisenberg model.

B. The evaluation of the loss integral

We now turn to the discussion of the loss integral, $I_{l_z}(L_1\sigma_1|L_2\sigma_2)$, Eq. (A12) of Appendix A. This term includes the derivative $[\partial v(\mathbf{r})/\partial S_{l_z}^-]_0^{\sigma\sigma'}$, which can be calculated by a reference to the single-site Hamiltonian used in the generation of the single-site t matrices.

In the case of spin-polarized electron scattering from magnetically ordered materials, the single-site Hamiltonian is given by[18]

$$H=c\boldsymbol{\alpha}\cdot\mathbf{p}+[\beta mc^2+V(\mathbf{r})]I_{4\times4}-\beta\boldsymbol{\sigma}\cdot\mathbf{B}(\mathbf{r})\ , \tag{23}$$

where $I_{4\times4}$ stands for the 4×4 identity matrix, $\boldsymbol{\alpha}$ and β follow the standard notation of Bjorken and Drell.[19] In this expression, $V(\mathbf{r})$ is the spin-averaged electrostatic potential, and $\mathbf{B}(\mathbf{r})$ is a measure of the exchange interaction.

In *ab initio* electronic structure calculations, such as those reported by Fu and Freeman[20] (whose potentials we have used in our calculations), the axis of quantization of the magnetic moments in the crystal are all taken parallel to each other, and to a particular direction we call $\hat{z}$. Thus, the potential which enters the Dirac equation, say for a single ion located at the origin of the coordinate system, is written $V(\mathbf{r})I_{4\times4}-\beta B(\mathbf{r})\sigma_z$. In our work, we extract from Fu and Freeman's full potentials only the spherically symmetric portion. From this, as mentioned before, we have obtained excellent accounts of SPLEED data on the Fe(110) surface over a wide energy range.[15,16] Thus, we write the potentials we use as $V(r)I_{4\times4}-\beta B(r)\sigma_z$.

Thermal fluctuations in the spin system lead to deviations in the directions of the spin $\mathbf{S}$ at the origin away from the $\hat{z}$ direction. We assume the exchange potential $B(r)$ just rotates rigidly with the spin. Thus, we replace the exchange potential $-\beta B(r)(\hat{z}\cdot\boldsymbol{\sigma})$ by the form $-\beta B(r)(\mathbf{S}\cdot\boldsymbol{\sigma})/S$, where we now regard the quantity $\mathbf{S}$ as an operator that operates on the eigenstates which describe the ground and excited states of the substrate spin system. The single-site Hamiltonian thus becomes

$$H=c\boldsymbol{\alpha}\cdot\mathbf{p}+\beta mc^2+V(r)-\beta\frac{B(r)}{S}\boldsymbol{\sigma}\cdot\mathbf{S}\ . \tag{24}$$

By introducing the usual ladder operators, $\sigma^{\pm}=\sigma_x\pm i\sigma_y$ and $S^{\pm}$, we obtain

$$H=c\boldsymbol{\alpha}\cdot\mathbf{p}+\beta mc^2+V(r)-\beta\frac{B(r)}{S}\sigma_zS_z -\beta\frac{B(r)}{2S}(\sigma^+S^-+\sigma^-S^+)\ . \tag{25}$$

The scattering potential associated with an ion core in a layer l_z can then be identified as

$$v_{l_z}(r)=V(r)-\beta\frac{B_{l_z}(r)}{S}\sigma_zS_z -\beta\frac{B_{l_z}(r)}{2S}(\sigma^+S^-+\sigma^-S^+)\ . \tag{26}$$

In a ferromagnet below the Curie temperature, all the moments are aligned, and we can take this direction of magnetization as the z axis of our coordinate system. At $T=0$ K, there are no thermal fluctuations of the magnetic moment, and we will have only the σ_zS_z term, but, as the temperature is increased, fluctuations around the magnetization direction start to occur. The last term of Eq. (26) represents the effects of these fluctuations.

We can now evaluate the derivative in $I_{l_z}(L_1\sigma_1|L_2\sigma_2)$. Since in our discussion of the scattering efficiency per unit solid angle we have worked with the large component of the Dirac four-spinor, in the above expression we replace the 4×4 matrix β by its upper block, the 2×2 identity matrix. The derivative is straightforward, and given by

$$\left[\frac{\partial v_{l_z}(r)}{\partial S^-_{l_z}}\right]_0=-\frac{B_{l_z}(r)}{2S}\sigma^+\ . \tag{27}$$

From this we see that (in the nonrelativistic limit) excitation of the substrate spin system is accompanied by a flip of the beam electron spin. For a relativistic electron, where the eigenstates are not eigenstates of σ_z, notice that spin excitations may be created in the substrate without a spin flip. However, the excitation probabilities will be small for the materials of interest here.

By using this result, we have the following for $I_{l_z}(L_1\sigma_1|L_2\sigma_2)$:

$$\begin{aligned}I_{l_z}(L_1\sigma_1|L_2\sigma_2)=&-\frac{1}{2S}i^{l_2-l_1}\\&\times\sum_{\sigma\sigma'}\int d\rho_3\rho_3^2d\Omega\Bigg[j_{l_1}(k\rho_3)Y_{l_1}^{m_1*}(\hat{\rho}_3)\delta_{\sigma,\sigma_1}\\&+\sum_{m_1'=-l_1}^{l_1}\Bigg[\int d\rho_1\rho_1^2d\rho_2\rho_2^2j_{l_1}(k\rho_1)\\&\times t^{\sigma_1\sigma}_{R_{z_{l_1}};m_1m_1'}(\rho_1,\rho_2)G^{\sigma}_{l_1}(\rho_2,\rho_3)\Bigg]Y_{l_1}^{m_1'*}(\hat{\rho}_3)\Bigg]\\&\times(\sigma^+)^{\sigma\sigma'}B_{l_z}(\rho_3)\Bigg[j_{l_2}(k\rho_3)Y_{l_2}^{m_2*}(\hat{\rho}_3)\delta_{\sigma',\sigma_2}+\sum_{m_2'=-l_2}^{l_2}\Bigg[\int d\rho_4\rho_4^2d\rho_5\rho_5^2G^{\sigma'}_{l_2}(\rho_3,\rho_4)\\&\times t^{\sigma'\sigma_2}_{R_{z_{l_2}};m_2m_2'}(\rho_4,\rho_5)j_{l_2}(k\rho_5)\Bigg]\\&\times Y_{l_2}^{m_2'}(\hat{\rho}_3)\Bigg]\ .\end{aligned} \tag{28}$$

The angular integration over $d\Omega_3$ can be done using the orthogonality of spherical harmonics. It gives $\delta_{l_1,l_2}\delta_{m_1',m_2'}$. We have

$$I_{l_z}(L_1\sigma_1|L_2\sigma_2)=-\frac{1}{2S}\delta_{l_1,l_2}i^{l_2-l_1}\sum_{\sigma\sigma'}\sum_{m_1'=-l_1}^{l_1}\int d\rho_3\rho_3^2\left[j_{l_1}(k\rho_3)\delta_{\sigma,\sigma_1}\delta_{m_1,m_1'}+\int d\rho_1\rho_1^2 d\rho_2\rho_2^2 j_{l_1}(k\rho_1)\times t^{\sigma_1\sigma}_{R_{z_{l_1}};m_1m_1'}(\rho_1,\rho_2)G^{\sigma}_{l_1}(\rho_2,\rho_3)\right]$$
$$\times(\sigma^+)^{\sigma\sigma'}B_{l_z}(\rho_3)\left[j_{l_1}(k\rho_3)\delta_{\sigma',\sigma_2}\delta_{m_2,m_1'}+\int d\rho_4\rho_4^2 d\rho_5\rho_5^2 G^{\sigma'}_{l_1}(\rho_3,\rho_4)\times t^{\sigma'\sigma_2}_{R_{z_{l_1}};m_1'm_2}(\rho_4,\rho_4)j_{l_1}(k\rho_5)\right]. \quad (29)$$

If we write

$$\psi^{(+)\sigma}_{\mathbf{k}(\text{scattered})}(\mathbf{r})=4\pi/\sqrt{V}\sum_{l,m,m'}a^{\sigma}_{l;m,m'}Y_l^m(\hat{r})Y_l^{m'}(\hat{k})R^{\sigma}_{l;m,m'}(r)\chi^{\sigma},$$

then, using the result of Appendix B, we can replace the large parentheses in Eq. (29) by the radial wave functions. The result can be written

$$I_{l_z}(L_1\sigma_1|L_2\sigma_2)=-\frac{1}{2S}\delta_{l_1,l_2}i^{l_2-l_1}\sum_{\sigma\sigma'}\sum_{m_1'=-l_1}^{l_1}\int d\rho\rho^2[\delta_{\sigma,\sigma_1}a^{\sigma}_{l_1;m_1,m_1'}R^{\sigma}_{l_1;m_1,m_1'}(\rho)]\times(\sigma^+)^{\sigma\sigma'}B_{l_z}(\rho)[\delta_{\sigma',\sigma_2}a^{\sigma'}_{l_1;m_1',m_2}R^{\sigma'}_{l_1;m_1',m_2}(\rho)]. \quad (30)$$

Within the muffin-tin model scheme, $B(\rho)$ can be taken to be zero outside the muffin-tin sphere. This is a good assumption since the $3d$ orbitals which contribute most to $B(\rho)$ are localized well inside the muffin-tin sphere, and $B(\rho)\to 0$ rapidly for $\rho\gtrsim R_{\text{MT}}$. Then, evaluation of the integral requires that the radial wave functions be known at all points inside the muffin-tin sphere. When we compute the single-site t matrices, we generate the radial functions in the course of the calculation as well. Thus, in principle, it is possible to evaluate this integral numerically. However, this would be time consuming. As a remedy, we approximate the Dirac Hamiltonian by the Pauli Hamiltonian *just* for the evaluation of this integral. This approximation will work very well for the $3d$ transition metals of interest here. This approximation allows one to phrase the result in terms of two separate integrations of the Schrödinger equation, one with the potential $V^{\uparrow}(r)=V(r)-B(r)$, the potential a spin-up electron experiences in the crystal, and another with $V^{\downarrow}(r)=V(r)+B(r)$. The full relativistic single-site t matrix, $t^{\sigma\sigma'}_{l;mm'}$ then reduces to t^{σ}_{l} which is diagonal in, and independent of, the azimuthal indices m and m', and which is characterized by the phase shifts $\delta_l^{\uparrow}$ ($\delta_l^{\downarrow}$) corresponding to the potential $V^{\uparrow}(r)$ [$V^{\downarrow}(r)$] through $e^{i\delta_l^{\sigma}}\sin(\delta_l^{\sigma})$. The coefficients $a^{\sigma}_{l;m,m'}$ simplify to $e^{i\delta_l^{\sigma}}$.[21] Note also that, since the m dependence of the t matrix is removed, the m_1' summation drops out.

From our definition of σ^+ above we see that $(\sigma^+)^{\sigma\sigma'}$ is nonzero only if ($\sigma=\uparrow$, $\sigma'=\downarrow$) holds. Thus, as remarked above, in the nonrelativistic limit, excitation of the substrate spin system is accompanied by a flip of the beam electron spin.

So we have

$$I_{l_z}(L_1\sigma_1|L_2\sigma_2)=-\frac{1}{2S}\delta_{l_1,l_2}\delta_{m_1,m_2}\delta_{\sigma_1,\uparrow}\delta_{\sigma_2,\downarrow}\times i^{l_2-l_1}e^{i\delta_{l_1}^{\uparrow(l_z)}}e^{i\delta_{l_1}^{\downarrow(l_z)}}\times 2\int_0^{R_{\text{MT}}}d\rho\rho^2R_{l_1}^{\uparrow}(\rho)\times B_{l_z}(\rho)R_{l_1}^{\downarrow}(\rho). \quad (31)$$

Here we choose to label the phase shifts by the layer index l_z because, in general, the top few layers of the surface region differ in magnetic moments from the bulk. The radial integral is evaluated in Appendix C, it is equal to $(1/2k)\sin[\delta_{l_1}^{\uparrow(l_z)}-\delta_{l_1}^{\downarrow(l_z)}]$. The final form of $I_{l_z}(L_1\sigma_1|L_2\sigma_2)$ is thus

$$I_{l_z}(L_1\sigma_1|L_2\sigma_2)=-\frac{1}{2S}\frac{1}{k}\delta_{L_1,L_2}\delta_{\sigma_1,\uparrow}\delta_{\sigma_2,\downarrow}e^{i\delta_{l_1}^{\uparrow(l_z)}+i\delta_{l_1}^{\downarrow(l_z)}}\times\sin[\delta_{l_1}^{\uparrow(l_z)}-\delta_{l_1}^{\downarrow(l_z)}]. \quad (32)$$

III. EXPLICIT CALCULATIONS OF SPIN-WAVE EXCITATION SCATTERING EFFICIENCIES

We now present results of explicit calculations of the scattering efficiency for exciting spin waves on the surfaces of ferromagnetic crystals using the formalism developed above. With the exception of the spin correlation functions $\langle S_+(\mathbf{q}_\parallel,l_z',t)S_-(\mathbf{q}_\parallel,l_z,0)\rangle$, all quantities which enter the final expression in Eq. (22) may be generated through use of the (relativistic) multiple-scattering theory developed by Feder,[22] and used in our earlier descriptions of spin-polarized elastic scattering from the Fe(110) surface.[14] This includes the matrix element which couples the beam electron to the spin excitation. We have seen in Appendix B that this may be expressed in terms of the phase shifts for scattering from a single muffin tin if we are willing to treat this object within the framework of nonrelativistic quantum mechanics.

We shall explore here the scattering efficiency for exciting spin waves on Fe and Ni surfaces, supposing these excitations may be described through use of a Heisenberg spin Hamiltonian. We generate the spin correlation functions of interest through use of spin-wave theory with frequencies and eigenvectors generated from a slab calculation. The procedure is quite similar to that employed in earlier studies of phonon excitation by low-energy electrons incident on surfaces;[9] by this means both surface spin-wave and bulk spin-wave contributions to the cross section are included. The earlier literature on phonon excitation by electrons establishes clearly that the electron excites both surface and bulk phonons and, as discussed in an early discussion of spin-wave excitations by electrons many years ago,[23] the same is true when electrons incident on surfaces excite spin waves. We direct the reader's attention to an early study of spin waves in a Heisenberg ferromagnetic slab, wherein the interplay between surface and bulk spin-wave contributions to surface phenomena is emphasized,[24] and a review article in which the properties of surface spin waves are reviewed, for localized spin models.[25]

We have used a model with nearest- and next-nearest-neighbor exchange interactions, with next-nearest-neighbor interactions half the strength of the nearest-neighbor coupling. This is roughly correct for Fe. The magnitudes are fitted to bulk spin-wave bandwidths estimated from the results of Ref. 13. At the moment, we have used bulk exchange constants near the surface since, at this time, we have little reliable information on hand on the magnitude of the effective exchange couplings near the surface. We note that it would be of very great interest to address this question theoretically, within the framework of *ab initio* ground-state calculations of magnetic materials. This could be done, at least in principle, by spin analogues of "frozen phonon" calculations that have been used to generate effective interatomic force constants near surfaces.

The description of spin-wave excitations near surfaces just described is quite crude, and thus of limited reliability, so far as reliable predictions of surface spin-wave dispersion relations and properties are concerned. The results, once again, are handicapped by our lack of knowledge of the effective exchange couplings near surfaces, and by the possibly substantial influence of damping by interaction of (short-wavelength) spin waves by coupling to Stoner excitations. Theses interactions damp bulk spin waves appreciably and modify their dispersion relation as well.[13] The primary aim of the present paper is to estimate, as quantitatively as possible, the scattering effficiency for exciting spin excitations at the surfaces of the much studied transition ferromagnets Fe and Ni. These overall scattering efficiencies [$d^2P/d\Omega\, d\omega$ in Eq. (22), integrated over energy loss $\hbar\omega$] will not be highly sensitive to the model used for the surface spin dynamics, since the overall scattering efficiency is easily seen to be related to static spin correlation functions, which are integrals over the entire frequency spectrum of spin fluctuations at the wave vector $\mathbf{q}_\parallel$ probed in the measurement. Our use of the specific model of spin excitations described in the previous paragraph will provide a notion of the range of energy loss of interest, and at least from the qualitative point of view, the features that can be expected in the loss spectrum.

After the exchange Hamiltonian for the model slab is chosen, we proceed by introducing the Holstein-Primakoff transformation. Thus, the operator $S_-(l_\parallel l_z)$ of Sec. II is written[24] with $a^\dagger(l_\parallel l_z)$ the boson creation operator in the site representation

$$S_-(l_\parallel,l_z)=\sqrt{2S}\,a^\dagger(l_\parallel l_z) = \left[\frac{2S}{N_s}\right]^{1/2}\sum_{\mathbf{q}_\parallel} a^\dagger(\mathbf{q}_\parallel,l_z)\exp[-i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel(l_\parallel)]\ . \tag{33}$$

The operator $S_-(\mathbf{q}_\parallel,l_z)$ is thus identified with the combination $(2S)^{1/2}a^\dagger(\mathbf{q}_\parallel l_z)$. We may let $a^\dagger(\mathbf{q}_\parallel,s)$ be the boson creation operator for creating the normal mode s of wave vector $\mathbf{q}_\parallel$. The index s may refer to either a surface spin wave or a bulk spin wave. One may introduce the eigenvector $e(\mathbf{q}_\parallel s,l_z)$ generated by diagonalizing the spin-wave Hamiltonian for the slab.[24] Then we have

$$a^\dagger(\mathbf{q}_\parallel,l_z)=\sum_s e(\mathbf{q}_\parallel s,l_z)a^\dagger(\mathbf{q}_\parallel s)\ . \tag{34}$$

where we assume the eigenvectors are normalized so that

$$\sum_{l_z}|e(\mathbf{q}_\parallel s,l_z)|^2=1\ . \tag{35}$$

The quantity $|e(\mathbf{q}_\parallel s,l_z)|^2$ measures the square of the amplitude of mode $(\mathbf{q}_\parallel s)$ in the lattice plane l_z.

The operator $a^\dagger(\mathbf{q}_\parallel s)$ has the time dependence $\exp[+i\omega(\mathbf{q}_\parallel s)t]$. Hence, in the spin-wave picture,

$$\int_{-\infty}^{+\infty}\frac{dt}{2\pi\hbar}\exp(i\omega_{if}t)\langle S_+(\mathbf{q}_\parallel,l_z',t)S_-(\mathbf{q}_\parallel,l_z,0)\rangle_T=\frac{2S}{\hbar}\sum_s e^*(\mathbf{q}_\parallel s,l_z')e(\mathbf{q}_\parallel s,l_z)[1+\bar{n}(\mathbf{q}_\parallel s)]\delta[\omega_{if}-\omega(\mathbf{q}_\parallel s)]\ . \tag{36}$$

Here

$$\bar{n}(\mathbf{q}_{\parallel}s)=\{\exp[\hbar\omega(\mathbf{q}_{\parallel}s)/k_BT]-1\}^{-1}$$

is the Bose-Einstein function. The sum over s in Eq. (26) includes all bulk spin waves associated with the wave vector $\mathbf{q}_{\parallel}$, along with surface spin waves. In practice, the Dirac δ function is replaced by a Lorentzian of finite width to simulate actual loss spectra.

For simple surfaces, and Heisenberg models such as those used here, it is also possible to generate analytic closed expressions for the required correlation functions through use of Green's-function methods.[23] This approach provides an alternate means of addressing aspects of the loss spectra, which can be particularly useful when subtle cancellations between bulk and spin-wave contributions to surface response characteristics are of interest.

In Fig. 2, for electrons scattered inelastically by spin-wave excitation from Fe(100), we show the form of the theoretical loss cross section, for a particular choice of scattering geometry. The incident energy is 100 eV, and the angle of incidence is 45°. The wave vector $\mathbf{q}_{\parallel}$ is directed along the line from $\bar{\Gamma}$ to $\bar{X}$ in the surface Brillouin zone. Here $\mathbf{q}_{\parallel}$ is 60% of the way to the zone boundary, from $\bar{\Gamma}$.

The prominent loss peak near 1000 cm^{-1} is provided by excitation of a surface spin wave. These modes lie below spin-wave band and thus are an analogue to the Rayleigh surface acoustical phonons of surface lattice dynamics (though when their wavelength is very long compared to a lattice constant, they penetrate far more deeply, and have frequencies very close to bulk spin waves which propagate parallel to the surface).[25] The loss band at higher frequencies, which terminates in an asymmetric peak near 3500 cm^{-1}, is produced by excitation of bulk spin waves by the beam electron. In this example, which is not atypical, the surface wave is the strongest feature in the loss spectrum.

We use as a measure of the intensity of the scattering efficiency per unit solid angle ($dP/d\Omega$), defined as

$$\frac{dP}{d\Omega}=\int_{-\infty}^{+\infty}d\omega\frac{d^2P}{d\Omega\,d\omega}\ . \tag{37}$$

The integral may be performed numerically from calculations such as those displayed in Fig. 2.

In Fig. 3, for scattering from Fe(100), and again with an angle of incidence of 45° and $\mathbf{q}_{\parallel}$ 60% of the way from $\bar{\Gamma}$ to $\bar{X}$, we show the magnitude and beam energy dependence of ($dP/d\Omega$). At nearly all energies, the surface spin wave is the strongest feature in the loss cross section, so to rather good approximation this may be viewed as the cross section for exciting the surface modes. We provide similar information for Ni(110) in Fig. 4. The angle of incidence is again 45° and the scattering geometry is such that $\mathbf{q}_{\parallel}$ is directed along the line from $\bar{\Gamma}$ to $\bar{X}$, with magnitude fixed to 60% of the distance out from $\bar{\Gamma}$. We see that, for exciting spin waves on the Ni surface, the excitation probability is about an order of magnitude smaller than for Fe, a result not unexpected from the smaller ground-state moment present in Ni. These calculations ignore the possibility that moments in the surface assume values larger than the bulk. Our experience with SPLEED calculations for Fe(110) suggests that the presence of enhanced surface moments will increase the excitation cross section only modestly, from 10 to 30%, depending on details of the scattering geometry.

We must now explore the significance of the magnitudes of ($dP/d\Omega$), displayed in the above figures. We argue that our calculated excitation cross sections are sufficiently large that spin waves can be probed by electron-energy-loss spectroscopy.

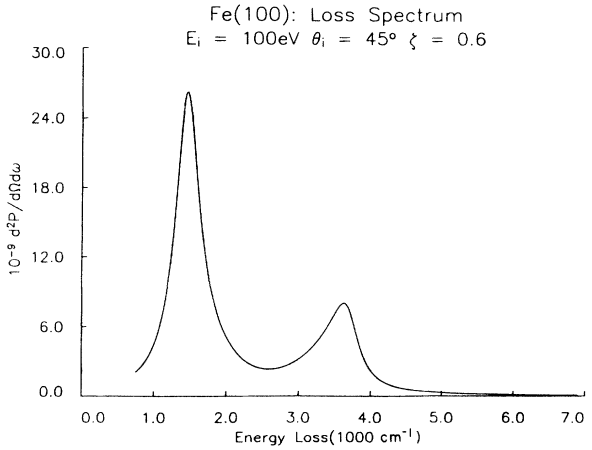

FIG. 2. The energy-loss spectrum calculated for our model of Fe(100). The electron energy is 100 eV, the angle of incidence is 45°, and the scattering plane is oriented so that the wave vector $\mathbf{q}_{\parallel}$ is directed along the line from $\bar{\Gamma}$ to $\bar{X}$. The loss spectrum is calculated for the case where $\mathbf{q}_{\parallel}$ is 60% of the way from $\bar{\Gamma}$ to $\bar{X}$. The prominent low-frequency loss peak is produced by excitation of a surface spin wave, and the higher-frequency band with a peak near 3500 cm^{-1} has its origin in the excitation of bulk spin waves.

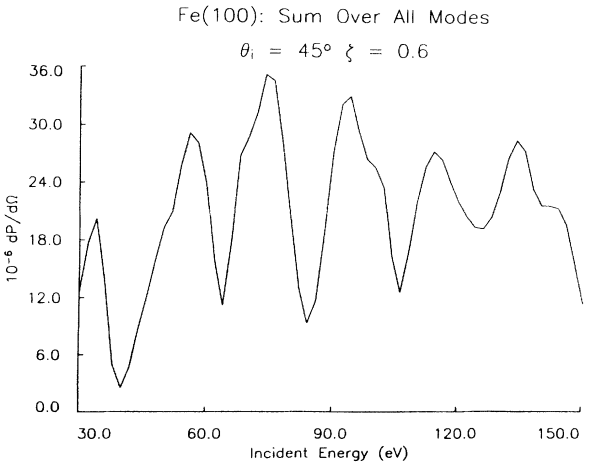

FIG. 3. The energy variation and magnitude of ($dP/d\Omega$) for exciting spin waves on Fe(100) for the scattering geometry employed in Fig. 2. The angle of incidence is 45°, and $\mathbf{q}_{\parallel}$ is directed along $\bar{\Gamma}$ to $\bar{X}$, with magnitude at each energy equal to 60% of the distance from $\bar{\Gamma}$ to $\bar{X}$. (The scattered beam direction thus varies with beam energy.)

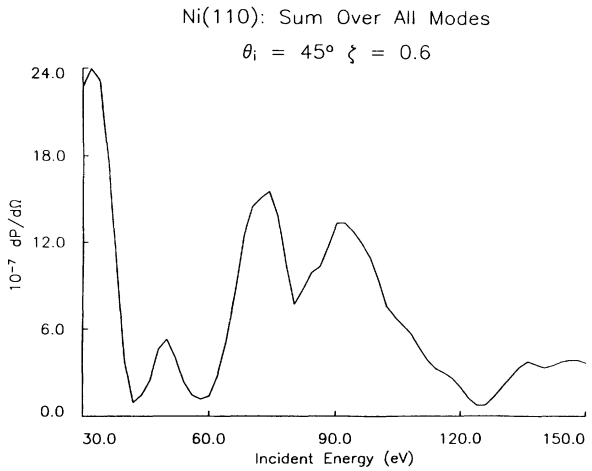

FIG. 4. The same as Fig. 3, but now for exciting spin waves on Ni(110). The angle of incidence is again 45°, with $\mathbf{q}_{\parallel}$ fixed to 60% of the distance from $\bar{\Gamma}$ to $\bar{X}$.

We may see this as follows. Suppose the number of electrons per unit time striking the surface is I_0, and the detector subtends solid angle $\Delta\Omega$. Then the number of electrons per unit time that strike the detector is $(dP/d\Omega)\Delta\Omega I_0$, if we include the total amount of scattering produced by all spin-wave modes which scatter electrons into $\Delta\Omega$. We envision an experiment similar to that reported by Abraham and Hopster,[4] wherein the incident beam is spin polarized, and the spin of the scattered electron is analyzed as well (the "complete experiment"). Such an experiment allows one to isolate contributions only from spin-flip processes. In Ref. 4, this approach was used to study the Stoner excitations on a Ni surface. Abraham and Hopster employed a beam current in the microamp range so I_0 is 10^{+13} electrons/sec. Their detector subtended an angle $\Delta\Theta \sim 2°$, so $\Delta\Omega = \pi(\Delta\Theta)^2 \simeq 10^{-3}$ s. We thus have, with $dP/d\Omega \sim 10^{-5}$ (for Fe, from Fig. 3), so roughly 10^5 electrons/sec reach the detector. The Mott detector used in this experiment has an efficiency in the range of 10^{-3}. Thus, if these conditions could be realized in a study of spin waves, the total spectrum would produce about 100 counts/sec for Fe, and an order of magnitude less for Ni (Fig. 4). The signals we estimate are very close to those realized in the experiments reported in Ref. 4.[26]

We conclude our calculated spin-wave cross sections fall very close to the scattering efficiencies realized in the earlier studies of Stoner excitations. If we were to have a complete theory of the spin excitations of itinerant ferromagnets such as Fe or Ni, the spin correlation functions which appear in Eq. (22) would contain contributions from *both* spin waves *and* Stoner excitations. Quite generally, such correlation functions are related to the appropriate dynamic susceptibility of the magnetic moment bearing electrons. In itinerant magnets, the dynamic susceptibilities contain poles produced by spin waves, which will give a contribution to the loss cross section, quite similar to that shown in Eq. (36), and branch cuts which, when their influence is incorporated, will describe the Stoner excitations.[27] Thus, cross sections for exciting Stoner excitations and for exciting spin waves should be close in value; the electron "sees" each excitation through the same spin correlation function. A task which remains for theory is a proper description of the dynamic susceptibility of an itinerant ferromagnet near its surface, within the framework of an analysis that includes both excitations.

The scattering cross sections for exciting spin excitations are very much smaller than for exciting phonons, in the off specular geometry used to study surface phonon dispersion relations by electron-energy-loss spectroscopy. We illustrate this in Fig. 5 where, for scattering from phonons on the Ni(110) surface, we show $(dP/d\Omega)$ as a function of beam energy. The scattering geometry is the same as that used in Figs. 3 and 4. We note the cross section for exciting phonons on the Fe and the Ni surface are very similar in magnitude. Finally, the reader will find detailed comparisons between energy-loss data on surface phonons and theory in earlier publications.[8]

Quite clearly, $(dP/d\Omega)$ associated with off specular phonon excitation is larger than that for exciting spin excitations and is roughly 3 orders of magnitude larger than for the spin excitations in the most favorable case we have explored, which is Fe. We have seen, however, that signals associated with spin excitation cross sections very close to those we calculate have been detected. The off specular phonon dispersion curve studies have utilized incident beam currents much smaller than those employed by Abraham and Hopster. The incident currents are in the range of 100 pA, rather than a μA. Also, to study the dispersion relation of surface phonons, one requires very high resolution in energy. Contemporary experiments employ resolution in the 3-meV range; Abraham and Hopster employed resolution of a few tens of meV (1 meV

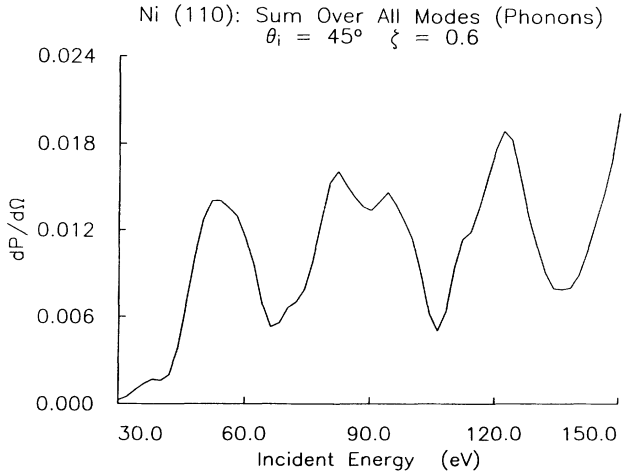

FIG. 5. A plot of the energy variation of $(dP/d\Omega)$, the probability of exciting all phonon modes on the surface, as a function of beam energy, for $\mathbf{q}_{\parallel}$ 60% of the way from $\bar{\Gamma}$ to $\bar{X}$, on the Ni(110) surface. The scattering geometry is the same as that used in Fig. 4.

$= 8\ \text{cm}^{-1}$). At least in a transition-metal ferromagnet such as Fe, as we see from Fig. 2, the spin-wave energies are very much higher than the phonon energies. Very high resolution will thus not be required, and this should also allow use of higher incident beam currents.[28]

We conclude with Fig. 6, in which we explore other isues. The first is the influence of alterations in exchange near the surface on the loss spectrum. In Fig. 6(a), we reproduce the loss spectrum in Fig. 2, and then in Fig. 6(b), we display the loss spectrum for the same scattering conditions, where the exchange couplings in the outermost surface layer are reduced from the bulk values by 50%. The surface spin wave is downshifted and the bulk spin-wave contributions to the spectrum reflect these changes only slightly.

We believe these should be of particular interest in the study of spin excitations of ultrathin films by the electron-energy-loss method. We show in Fig. 6(c) a loss spectrum of a three-layer Fe(100) film, with exchange couplings equal to their bulk value everywhere. For each value of $\mathbf{q}_{\parallel}$, we now have three spin-wave modes, which are standing wave excitations of the film. The spin-wave eigenvectors are entirely confined to the ultrathin film, if this film is placed on a nonmagnetic substrate. The value of $(dP/d\Omega)$ calculated for the three-layer film (the integrated strength of the loss cross section) differs little from that for semi-infinite Fe. Thus, ultrathin films can be explored as easily as thick samples with this method. We show in Fig. 6(d) the loss spectrum calculated for a three-layer film with exchange couplings in the two outer layers reduced from the bulk values by 50%. The downshift in frequency of the various features in the spectrum is evident.

It is our hope that the calculations presented here will stimulate new experimental studies of spin excitations at ferromagnetic surfaces and in ultrathin films. At the moment, in our view, our lack of understanding of the nature of the short-wavelength spin excitation spectrum of these systems places severe limits on our ability to understand their basic properties.

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APPENDIX A

In the text we have defined a coefficient $A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)$ as

$$A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)=\int d^3r_1 d^3r_2 \exp(-i\mathbf{k}\cdot\mathbf{r}_1) \times\left[\frac{\partial T(\mathbf{r}_1\mathbf{r}_2;\{\mathbf{R}(l)\},\{\mathbf{S}(l)\})}{\partial S_-(l_z)}\right]_0^{\sigma\sigma_i} \times\exp(i\mathbf{k}_i\cdot\mathbf{r}_2)\ , \tag{A1}$$

which is the matrix element that links the incident plane wave to the outgoing scattered wave. In the following, we obtain an explicit representation for this matrix element. The discussion here is phrased in language appropriate to the relativistic calculations that we have used in recent work.[15,16] Additionally, the following analysis is carried out within the framework of standard multiple-scattering theory, which we review for completeness.

From the definition of T,[16] we have

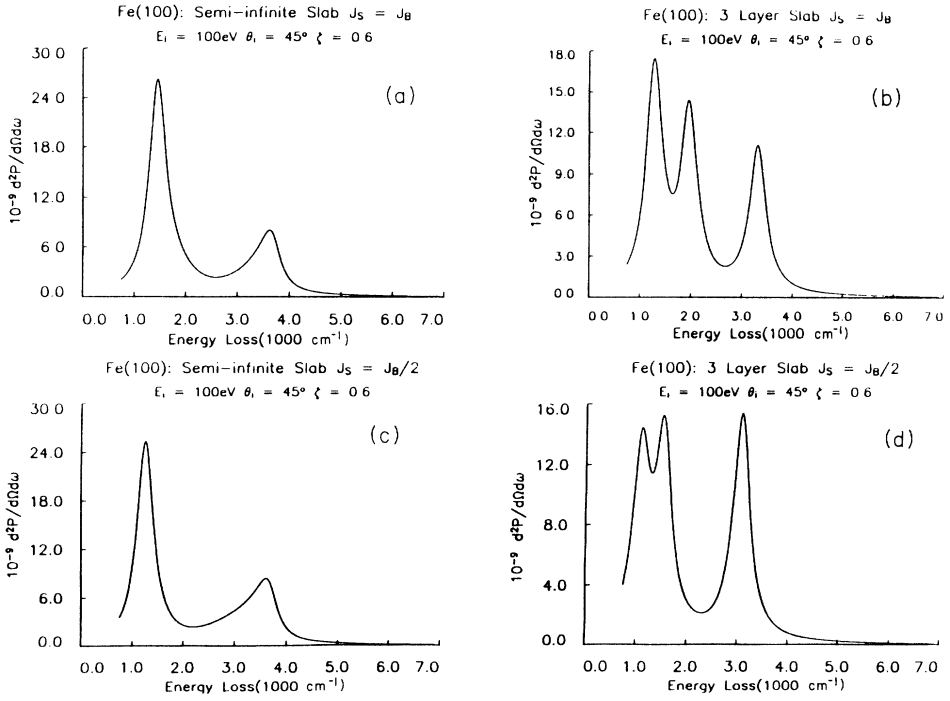

FIG. 6. The energy-loss spectrum for (a) the semi-infinite Fe crystal with (100) surfaces, and all exhange interactions equal to those in the bulk (this is the same loss spectrum given in Fig. 2), (b) the semi-infinite Fe(100) crystal, where all exchange interactions within the surface are reduced 50% from the bulk values, (c) a three-layer Fe film, with all exchange couplings equal to the bulk, and (d) a three-layer Fe film with exchange interactions in the outer layers reduced by 50% from the bulk values.

$$T=V+VGT\ , \tag{A2}$$

so that

$$\left[\frac{\partial T}{\partial S_-(l_\parallel=0,1_z)}\right]_0 \equiv \left[\frac{\partial T}{\partial S_-(l_z)}\right]_0 = \left[(1-VG)^{-1}\left[\frac{\partial V}{\partial S_-(l_z)}\right]_0 \times(1+GT)\right]_0 , \tag{A3}$$

which can easily be shown to be identical to

$$\left[\frac{\partial T}{\partial S_-(l_z)}\right]_0 =(1+T_0G)\left[\frac{\partial V}{\partial S_-(l_z)}\right]_0(1+GT_0)\ . \tag{A4}$$

In order to continue on, we need to express T_0, the entire crystal scattering T matrix, in terms of $t_{R(l)}$, the single-site scattering T matrix. For multiple scattering from a single-site scattering we have

$$t_R=v_R+v_RGt_R \tag{A5}$$

and so, for T_0, we obtain[16]

$$T_0=\sum_{R_1}t_{R_1}+\sum_{R_1,R_2}{}'\ t_{R_1}Gt_{R_2} + \sum_{R_1,R_2,R_3}{}'\ t_{R_1}Gt_{R_2}Gt_{R_3}+\cdots\ , \tag{A6}$$

where the prime over summation signs indicate that consecutive R_i are not to be equal [e.g., $\sum'_{R_1,R_2,R_3}=\sum_{R_1,R_2,R_3}(R_1\neq R_2,R_2\neq R_3)$, etc]. Thus, if we insert this expression for T_0 into $[\partial T/\partial S_-(l_z)]_0$ in $A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)$ then, in coordinate and spin space, we obtain

$$\begin{aligned}A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)=&\sum_{\sigma_1\cdots\sigma_4}\int d^3r_1\cdots d^3r_9\exp[-i(\mathbf{k}\cdot\mathbf{r}_1)]\\ &\times\left[\delta(\mathbf{r}_1-\mathbf{r}_2)\delta_{\sigma\sigma_1}+\sum_{R_1\neq R_z}t_{R_1}^{\sigma\sigma_1}(\mathbf{r}_1-\mathbf{R}_1,\mathbf{r}_2-\mathbf{R}_1)G^{\sigma_1}(\mathbf{r}_2-\mathbf{r}_3)+\cdots\right]\\ &\times[\delta(\mathbf{r}_3-\mathbf{r}_5)\delta_{\sigma_1\sigma_2}+t_{R_z}^{\sigma_1\sigma_2}(\mathbf{r}_3-\mathbf{R}_z,\mathbf{r}_4-\mathbf{R}_z)G^{\sigma_2}(\mathbf{r}_4-\mathbf{r}_5)]\\ &\times\left[\frac{\partial v_{l_z}(\mathbf{r}_5)}{\partial S_-(l_z)}\right]_0^{\sigma_2\sigma_3}[\delta(\mathbf{r}_5-\mathbf{r}_7)\delta_{\sigma_3\sigma_4}+G^{\sigma_3}(\mathbf{r}_5-\mathbf{r}_6)+t_{R_z}^{\sigma_3\sigma_4}(\mathbf{r}_6-\mathbf{R}_z,\mathbf{r}_7-\mathbf{R}_z)]\\ &\times\left[\delta(\mathbf{r}_7-\mathbf{r}_9)\delta_{\sigma_4\sigma_i}\sum_{R_1\neq R_z}G^{\sigma_4}(\mathbf{r}_7-\mathbf{r}_8)t_{R_1}^{\sigma_4\sigma_i}(\mathbf{r}_8-\mathbf{R}_1,\mathbf{r}_9-\mathbf{R}_1)+\cdots\right]\exp(-i\mathbf{k}_i\cdot\mathbf{r}_9)\ .\end{aligned} \tag{A7}$$

Note we now use R_z to denote $\mathbf{R}(l_\parallel=0,l_z)$. If we now shift the arguments of the t matrices such that $\mathbf{r}_i\rightarrow\boldsymbol{\rho}_i+\mathbf{R}_i$ and use the following expansions,[16]

$$t_R^{\sigma\sigma'}=\sum_{LL'}\delta_{ll'}t_{R_{LL'}}^{\sigma\sigma'}(\rho\rho')Y_L(\hat{\rho})Y_{L'}^*(\hat{\rho}')\ , \tag{A8}$$

$$\exp(i\mathbf{k}\cdot\boldsymbol{\rho})=4\pi\sum_L i^l j_l(k\rho)Y_L(\hat{\rho})Y_L^*(\hat{k})\ , \tag{A9}$$

$$G^\sigma(\boldsymbol{\rho}'-\boldsymbol{\rho}+\mathbf{R}_1-\mathbf{R}_2)=-(4\pi ik_i)\frac{2\pi}{\hbar^2}F^\sigma(k_i)\sum_{L_1L_2L_3}h_{l_3}^{(1)}(k_i|\mathbf{R}_1-\mathbf{R}_2|)j_{l_1}(k_i\rho')j_{l_2}(k_i\rho)i^{l_1-l_2+l_3}\times a(L_1L_2L_3)Y_{L_1}(\hat{\rho}')Y_{L_2}^*(\hat{\rho})Y_{L_3}(\mathbf{R}_1-\mathbf{R}_2)\ , \tag{A10}$$

where L denotes the pair (l,m), j and $h^{(1)}$ are the spherical Bessel and Hankel functions,

$$a(L_1L_2L_3)\equiv\int d\Omega(\hat{k})Y_{L_1}^*(\hat{k})Y_{L_2}(\hat{k})Y_{L_3}^*(\hat{k})\ ,$$

and

$$F^\sigma(k_i)=2k_i\Big/\left[2k_i+\frac{2m}{\hbar^2}\left[\frac{\partial\Sigma^\sigma(k,E)}{\partial k}\right]_{k_i}\right]\ ,$$

then the expression for $A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)$ becomes

$$A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)=-(4\pi)^2\sum_{LL'}\sum_{L_1L_2}\sum_{\sigma_1\sigma_2}Y_L(\hat{k})\left[\delta_{\sigma\sigma_1}\delta_{LL_1}+\sum_{R_1\neq R_z}[t_{R_1}^{\sigma\sigma_1}(k)G^{\sigma_1}(\mathbf{R}_1-\mathbf{R}_z)]_{LL_1}\exp[-i\mathbf{k}\cdot(\mathbf{R}_1-\mathbf{R}_z)]+\cdots\right]$$
$$\times\exp(-i\mathbf{k}\cdot\mathbf{R}_z)I_{l_z}(L_1\sigma_1|L_2\sigma_2)\exp(i\mathbf{k}_i\cdot\mathbf{R}_z)$$
$$\times\left[\delta_{\sigma_2\sigma_1}\delta_{L_2L'}+\sum_{R_1\neq R_z}[G^{\sigma_2}(\mathbf{R}_1-\mathbf{R}_z)t_{R_1}^{\sigma_2\sigma_i}(k_i)]_{L_2L'}\right.$$
$$\left.\times\exp[i\mathbf{k}_i\cdot(\mathbf{R}_1-\mathbf{R}_z)]+\cdots\right]Y_{L'}^*(\hat{k}_i)\ , \tag{A11}$$

where

$$G_{LL'}^{\sigma}(\mathbf{R}_1-\mathbf{R}_2)=\left[\frac{8\pi mk_i}{i\hbar^2}\right]\sum_{L''}i^{l''}a(LL'L'')h_{l''}^{(1)}(k_i|\mathbf{R}_1-\mathbf{R}_2|)Y_{L''}(\mathbf{R}_1-\mathbf{R}_2)i^{l-l'}\ ,$$

$$t_{RLL'}^{\sigma\sigma'}(k)=\delta_{ll'}\int d\rho_1 d\rho_2\rho_1^2\rho_2^2 j_l(k\rho_1)t_{RLL'}^{\sigma\sigma'}(\rho_1\rho_2)j_l(k\rho_2)\ ,$$

and we have used matrix notation to express terms of the form $\sum_{L''}t_{R_LL''}^{\sigma\sigma'}(k_i)G_{L''L'}^{\sigma'}(\mathbf{R}_1-\mathbf{R}_2)$ as $[t_R^{\sigma\sigma'}(k_i)G^{\sigma'}(\mathbf{R}_1-\mathbf{R}_2)]_{LL'}$. Also, in the above equation for $A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)$, we have made the definition

$$I_{l_z}(L_1\sigma_1|L_2\sigma_2)=i^{-(l_1-l_2)}\sum_{\sigma\sigma'}\int d^3\rho_1\cdots d^3\rho_5 j_{l_1}(k_i\rho_1)Y_{L_1}^*(\hat{\rho}_1)[\delta_{\sigma_1\sigma}\delta(\boldsymbol{\rho}_1-\boldsymbol{\rho}_2)+t_{R_z}^{\sigma_1\sigma}(\boldsymbol{\rho}_1,\boldsymbol{\rho}_3)G^{\sigma}(\boldsymbol{\rho}_3-\boldsymbol{\rho}_2)]$$
$$\times\left[\frac{\partial v_{l_z}(\boldsymbol{\rho}_2-\mathbf{R}_z)}{\partial S_-(l_z)}\right]_0^{\sigma\sigma'}[\delta_{\sigma'\sigma_2}\delta(\boldsymbol{\rho}_2-\boldsymbol{\rho}_4)+G^{\sigma'}(\boldsymbol{\rho}_2-\boldsymbol{\rho}_5)t_{R_z}^{\sigma'\sigma_2}(\boldsymbol{\rho}_5,\boldsymbol{\rho}_4)]$$
$$\times j_{l_2}(k_i\rho_4)Y_{L_2}(\hat{\rho}_4)\ . \tag{A12}$$

In essence, this is the matrix element of the potential through which the electron senses the spin excitation, evaluated between the initial- and final-state electron partial waves, associated with the angular momenta and spin indicies indicated.

Now, in the following, we shall evaluate $A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)$ and obtain a master formula for the scattering efficiency per unit solid angle per loss energy in terms of I_{l_z} since I_{l_z} is model specific. After having obtained the master formula, we shall apply it to the case in which $[\partial v_{l_z}(\mathbf{r})/\partial S_-(l_z)]_0$ is spherically symmetric. So, with this in mind, we continue on to evaluate the infinite lattice sums which appear in the bracketed factors of $A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)$. To facilitate the evaluation we make the following definitions:

$$J_N^z=\exp(-i\mathbf{k}_i\cdot\mathbf{R}_z)\sum_{\substack{R_1\cdots R_N\\R_1\neq R_z}}{}'G(\mathbf{R}_z-\mathbf{R}_1)t_{R_1}(k_i)\times\cdots\times G(\mathbf{R}_{N-1}-\mathbf{R}_N)t_{R_N}(k_i)\exp(i\mathbf{k}_i\cdot\mathbf{R}_N)\ ,$$
$$S_N^z=\sum_{\substack{R_1\cdots R_N\\R_N\neq R_z}}{}'\exp(-i\mathbf{k}\cdot\mathbf{R}_1)t_{R_1}(k)G(\mathbf{R}_1-\mathbf{R}_2)\times\cdots\times t_{R_N}(k)G(\mathbf{R}_N-\mathbf{R}_z)\exp(i\mathbf{k}\cdot\mathbf{R}_z)\ . \tag{A13}$$

Note, in these equations we have let G and t be 2×2 matrices with $(G)_{\sigma\sigma'}=\delta_{\sigma\sigma'}G^{\sigma}$. With these definitions it is evident that $A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)$ may be expressed as

$$A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)=-(4\pi)^2\sum_{LL'}\sum_{L_1L_2}\sum_{\sigma_1\sigma_2}Y_L(\hat{k})\left[\sum_{N=0}^{\infty}S_N^z\right]_{L\sigma,L_1\sigma_1}\exp(-i\mathbf{k}\cdot\mathbf{R}_z)$$
$$\times I_{l_z}(L_1\sigma_1|L_2\sigma_2)\exp(i\mathbf{k}_i\cdot\mathbf{R}_z)\left[\sum_{N=0}^{\infty}J_N^z\right]_{L_2\sigma_2,L'\sigma_i}Y_{L'}^*(\hat{k}_i)\ . \tag{A14}$$

Now, in the expressions for S_N^z and J_N^z, if we let $\mathbf{R}_i\rightarrow\mathbf{d}_{z_i}+\mathbf{P}_{z_i}$ where $\mathbf{d}_{z_i}$ denotes a vector from some global origin to an origin in the layer at l_{z_i} and $\mathbf{P}_{z_i}$ denotes a two-dimensional lattice vector in this layer, and if we note that, for a given layer, all single-site t matrices are equal, then we obtain

$$J_N^z=\sum_{z_1\cdots z_N}[\delta_{zz_1}G^{z_1z_1}t_{z_1}+(1-\delta_{zz_1})G^{zz_1}t_{z_1}]\times\cdots\times(\delta_{z_{N-1}z_N}G^{z_Nz_N}t_{z_N}+(1-\delta_{z_{N-1}z_N})G^{z_{N-1}z_N}t_{z_N}) ,$$
$$S_N^z=\sum_{z_1\cdots z_N}[\delta_{z_1z_2}t_{z_1}G^{z_1z_1}+(1-\delta_{z_1z_2})t_{z_1}G^{z_1z_2}]\times\cdots\times[\delta_{z_Nz}t_{z_N}G^{z_Nz_N}+(1-\delta_{z_Nz})t_{z_n}G^{z_Nz}] , \tag{A15}$$

where we have used z_i to denote l_{z_i}, supressed the k dependences, and made the definitions

$$G^{z_iz_i}(\mathbf{k})=\sum_{\mathbf{P}_{z_i}}\exp(-i\mathbf{k}\cdot\mathbf{P}_{z_i})G(\mathbf{P}_{z_i}) ,$$
$$G^{z_iz_j}(\mathbf{k})=(1-\delta_{z_iz_j})\sum_{\mathbf{P}_{z_i}\mathbf{P}_{z_j}}\exp[-i\mathbf{k}\cdot(\mathbf{d}_{z_i}-\mathbf{d}_{z_j}+\mathbf{P}_{z_i}-\mathbf{P}_{z_j})]G(\mathbf{d}_{z_i}-\mathbf{d}_{z_j}+\mathbf{P}_{z_i}-\mathbf{P}_{z_j}) . \tag{A16}$$

If we multiply through by the first factor of J_N^z and the last factor of S_N^z, sum the result from $N=0$ to $N\infty$, and let $\sum_{N=0}^{\infty}J_N^z=J^z$, $\sum_{N=0}^{\infty}S_N^z=S^z$, then we obtain the relations

$$J^z=(1-G^{zz}t_z)^{-1}\left[1+\sum_{z_1}G^{zz_1}t_{z_1}J^{z_1}\right] , \quad S^z=\left[1+\sum_{z_1}S^{z_1}t_{z_1}G^{z_1z}\right](1-t_zG^{zz})^{-1} . \tag{A17}$$

Now, we need to solve these equations for J^z and S^z. To this end, we use matrix notation and define certain vectors and matrices whose elements are indexed by *layers* as follows:

$$(J_1)_i=J^i , \quad (J_2)_i=t_iJ^i , \quad (S_1)_i=S^i , \quad (S_2)_i=S^it_i , \quad (R)_i=1 , \quad (G_{ij})=(1-\delta_{ij})G^{ij} , \quad (\tau)_{ij}=\delta_{ij}t_i(1-G^{ii}t_i)^{-1} . \tag{A18}$$

With these definitions we can obtain the following matrix equations for J_2 and S_2:

$$J_2=\tau(R+GJ_2) , \quad \tilde{S}_2=(\tilde{R}+\tilde{S}_2G)\tau , \tag{A19}$$

where "~" denotes transpose. Solving these equations, we find J^z and S^z to be given by

$$J^z=\sum_{z_1}(1-G^{zz}t_z)^{-1}(1-G\tau)^{-1}_{zz_1} , \quad S^z=\sum_{z_1}(1-\tau G)^{-1}_{z_1z}(1-t_zG^{zz})^{-1} . \tag{A20}$$

Thus, going back to the equation for $A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)$, we find

$$A_{l_z}(\mathbf{k}\sigma,\mathbf{k}_i\sigma_i)=-(4\pi)^2\sum_{LL'}Y_L(\hat{k})\sum_{z_1z_2}\{[1-\tau(\mathbf{k})G(\mathbf{k})]^{-1}_{z_1z}\exp(-i\mathbf{k}\cdot\mathbf{R}_z)$$
$$\times Q(l_z)\exp(i\mathbf{k}_i\cdot\mathbf{R}_z)[1-G(\mathbf{k}_i)\tau(\mathbf{k}_i)]^{-1}_{zz_2}\}_{L\sigma,L'\sigma_i}Y_L^*(\hat{k}_i) , \tag{A21}$$

where we have defined $Q(l_z)=(1-t_zG^{zz})^{-1}I_{l_z}(1-G^{zz}t_z)^{-1}$.

APPENDIX B

When one sets out to solve the Dirac equation, the wave function is expanded in terms of the spin-angle functions which are eigenfunctions of both J_z, the z component of the total angular momentum, and the operator $K=\beta(I+\boldsymbol{\sigma}\cdot\mathbf{L}/\hbar)$. In Sec. II, where we are computing the scattering efficiency per unit solid angle per energy loss, however, we use eigenfunctions of the orbital angular momentum, spherical Harmonics, and Pauli spinors as the basis of various expansions. Therefore, in order to simplify the expressions appearing in large parentheses in Eq. (29) of Sec. II, we consider an expansion of the scattered portion of the wave function in $(l,m;\text{spin})$ basis of the form

$$\psi_{\mathbf{k}}^{(+)\sigma}(\mathbf{r})=\frac{4\pi}{\sqrt{V}}\sum_{l,m,m'}a^{\sigma}_{l;m,m'}Y_l^m(\hat{r})Y_l^{m'*}(\hat{k})R^{\sigma}_{l;m,m'}(r)\chi^{\sigma} . \tag{B1}$$

The Lippmann-Schwinger equation reads

$$\psi_{\mathbf{k}}^{(+)\sigma}(\mathbf{r})=\phi_{\mathbf{k}}^{\sigma}(\mathbf{r})+\sum_{\sigma'=\uparrow,\downarrow}\int d^3r'G^{\sigma}(\mathbf{r}-\mathbf{r}')V^{\sigma,\sigma'}(\mathbf{r}')\psi_{\mathbf{k}}^{(+)\sigma'}(\mathbf{r}')$$
$$=\phi_{\mathbf{k}}^{\sigma}(\mathbf{r})+\sum_{\sigma'=\uparrow,\downarrow}\int\int d^3r'd^3r''G^{\sigma}(\mathbf{r}-\mathbf{r}')T^{\sigma,\sigma'}(\mathbf{r}',\mathbf{r}'')\phi_{\mathbf{k}}^{\sigma'}(\mathbf{r}'') . \tag{B2}$$

Here, $\phi_{\mathbf{k}}^{\sigma}(\mathbf{r})$ is the wave function for the incident electron, $G^{\sigma}(\mathbf{r}-\mathbf{r}')$ is the Green's function, $V^{\sigma,\sigma'}(\mathbf{r})$ is the scattering potential, and $T^{\sigma,\sigma'}(\mathbf{r}',\mathbf{r}'')$ is the corresponding t matrix. We introduce the following expansions:

$$\phi_{\mathbf{k}}^{\sigma}(\mathbf{r})=\chi^{\sigma}\frac{4\pi}{\sqrt{V}}\sum_{L}i^{l}j_{l}(kr)Y_{L}^{*}(\hat{k})Y_{L}(\hat{r})\ , \tag{B3}$$

$$G^{\sigma}(\mathbf{r}-\mathbf{r}')=\sum_{L}G_{l}^{\sigma}(r,r')Y_{L}^{*}(\hat{r}')Y_{L}(\hat{r})\ , \tag{B4}$$

$$T^{\sigma,\sigma'}(\mathbf{r}',\mathbf{r}'')=\sum_{l}\sum_{m,m'}t_{l;mm'}^{\sigma,\sigma'}(r',r'')Y_{l}^{m'*}(\hat{r}'')Y_{l}^{m}(\hat{r}')\ . \tag{B5}$$

We insert these expansions into the Lipmann-Schwinger equation (B2). Affecting the integrals over the solid angles, we obtain the following for the right-hand side:

$$\text{rhs}=\frac{4\pi}{\sqrt{V}}\sum_{l,m}i^{l}Y_{l}^{m}(\hat{r})\left[j_{l}(kr)Y_{l}^{m*}(\hat{k})\chi^{\sigma}+\sum_{\sigma'}\sum_{m'}\left[\int dr'r'^{2}dr''r''^{2}G_{l}^{\sigma}(r,r')t_{l;mm'}^{\sigma,\sigma'}(r',r'')j_{l}(kr'')\right]\chi^{\sigma'}Y_{l}^{m'*}(\hat{k})\right]\ , \tag{B6}$$

while the left-hand side is given by Eq. (B1).

Now, we multiply both sides by $Y_{l_1}^{m_1*}(\hat{r})$ and integrate over $d\Omega_{\hat{r}}$. We get

$$\chi^{\sigma}\sum_{m'}a_{l;m,m'}^{\sigma}Y_{l}^{m'*}(\hat{k})R_{l;m,m'}^{\sigma}(r)=i^{l}\sum_{m'}Y_{l}^{m'*}(\hat{k})\left[j_{l}(kr)\delta_{m,m'}\chi^{\sigma}+\sum_{\sigma'}\left[\int dr'r'^{2}dr''r''^{2}G_{l}^{\sigma}(r,r')t_{l;mm'}^{\sigma,\sigma'}(r',r'')j_{l}(kr'')\right]\chi^{\sigma'}\right]\ . \tag{B7}$$

Then, we repeat the above procedure by using $Y_{l_1}^{m_1}(\hat{k})$ and end up with

$$\chi^{\sigma}a_{l;m,m'}^{\sigma}R_{l;m,m'}^{\sigma}(r)=i^{l}\left[j_{l}(kr)\delta_{m,m'}\chi^{\sigma}+\sum_{\sigma'}\left[\int dr'r'^{2}dr''r''^{2}G_{l}^{\sigma}(r,r')t_{l;mm'}^{\sigma,\sigma'}(r',r'')j_{l}(kr'')\right]\chi^{\sigma'}\right]\ , \tag{B8}$$

or, equivalently,

$$\sum_{\sigma'}\delta_{\sigma,\sigma'}\chi^{\sigma'}a_{l;m,m'}^{\sigma'}R_{l;m,m'}^{\sigma'}(r)=i^{l}\sum_{\sigma'}\left[j_{l}(kr)\delta_{m,m'}\delta_{\sigma,\sigma'}+\left[\int dr'r'^{2}dr''r''^{2}G_{l}^{\sigma}(r,r')t_{l;mm'}^{\sigma,\sigma'}(r',r'')j_{l}(kr'')\right]\right]\chi^{\sigma'}\ . \tag{B9}$$

Equating the coefficients of $\chi^{\sigma'}$ on both sides, we obtain the result

$$\delta_{\sigma,\sigma'}a_{l;m,m'}^{\sigma'}R_{l;m,m'}^{\sigma'}(r)=i^{l}\left[j_{l}(kr)\delta_{m,m'}\delta_{\sigma,\sigma'}+\int dr'r'^{2}dr''r''^{2}G_{l}^{\sigma}(r,r')t_{l;mm'}^{\sigma,\sigma'}(r',r'')j_{l}(kr'')\right]\ . \tag{B10}$$

We can absorb the i^{-l} factor into the coefficients $a_{l;m,m'}^{\sigma'}$. The right-hand side is then exactly what we have in the second large parentheses of Eq. (29) of the text. A similar argument will enable us to replace the first large parentheses of Eq. (29) similarly:

$$\delta_{\sigma,\sigma'}a_{l;m,m'}^{\sigma'}R_{l;m,m'}^{\sigma'}(r)=i^{l}\left[j_{l}(kr)\delta_{m,m'}\delta_{\sigma,\sigma'}+\int dr'r'^{2}dr''r''^{2}j_{l}(kr')t_{l;mm'}^{\sigma,\sigma'}(r',r'')G_{l}^{\sigma'}(r',r'')\right]\ . \tag{B11}$$

APPENDIX C

In Eq. (31) of Sec. II, we have come across the following radial integral:

$$I=\int_{a}^{R_{\text{MT}}}dr\,r^{2}R_{l}^{\uparrow}(r)B(r)R_{l}^{\downarrow}(r)\ . \tag{C1}$$

From the definitions introduced in the main text, we have $B=\frac{1}{2}(V^{\downarrow}-V^{\uparrow})$, and $V=\frac{1}{2}(V^{\downarrow}+V^{\uparrow})$. The Schrödinger equations for spin-up and spin-down electrons are

$$\left[-\frac{\hbar^{2}}{2m}\nabla^{2}+V^{\uparrow}(r)\right]\psi^{\uparrow}(\mathbf{r})=E\psi^{\uparrow}(\mathbf{r})\ ,$$

$$\left[-\frac{\hbar^{2}}{2m}\nabla^{2}+V^{\downarrow}(r)\right]\psi^{\downarrow}(\mathbf{r})=E\psi^{\downarrow}(\mathbf{r})\ .$$

These will result in the following radial equations, after converting to atomic units:

$$-\ddot{u}_{l}^{\uparrow}+\frac{l(l+1)}{r^{2}}u_{l}^{\uparrow}+V^{\uparrow}u_{l}^{\uparrow}=Eu_{l}^{\uparrow}\ , \tag{C2}$$

$$-\ddot{u}_{l}^{\downarrow}+\frac{l(l+1)}{r^{2}}u_{l}^{\downarrow}+V^{\downarrow}u_{l}^{\downarrow}=Eu_{l}^{\downarrow}\ , \tag{C3}$$

where $u_{l}^{\sigma}(r)\equiv rR_{l}^{\sigma}(r)$, $\sigma=\uparrow,\downarrow$, and the overdots denote differentiation with respect to the reduced radial coordinates. We multiply Eq. (C2) by $u_{l}^{\downarrow}$, Eq. (C3) by $u_{l}^{\uparrow}$, and take their difference:

$$u_{l}^{\downarrow}\ddot{u}_{l}^{\uparrow}-u_{l}^{\uparrow}\ddot{u}_{l}^{\downarrow}+2u_{l}^{\uparrow}B(r)u_{l}^{\downarrow}=0\ . \tag{C4}$$

We integrate Eq. (C4) from zero to R_{MT}:

$$I=\tfrac{1}{2}\int dr\left\{\frac{d}{dr}(u_l^{\uparrow}\dot{u}_l^{\downarrow}-u_l^{\downarrow}\dot{u}_l^{\uparrow})\right\}$$
$$=\tfrac{1}{2}[u_l^{\uparrow}\dot{u}_l^{\downarrow}-u_l^{\downarrow}\dot{u}_l^{\uparrow}]_{R_{\mathrm{MT}}} \quad \text{(C5)}$$

since u_l^{σ} are zero at the origin. At $r=R_{\mathrm{MT}}$ we can use the asymptotic wave function for u_l^{σ}'s since the wave function is continuous across the muffin-tin sphere. We have

$$u_l^{\sigma}(r)=[j_l(kr)\cos\delta_l^{\sigma}-\eta_l(kr)\sin\delta_l^{\sigma}]r \quad \text{(C6)}$$

for $r\geq R_{\mathrm{MT}}$. Evaluating the Wronskian appearing in Eq. (C5) by using the above expression for u_l^{σ}, we end up with

$$I=\frac{1}{2k}\sin(\delta_l^{\uparrow}-\delta_l^{\downarrow})\ . \quad \text{(C7)}$$

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