Electron energy loss by electron-hole excitations in ferromagnets: The near-specular geometry

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A low-energy electron which reflects off a crystal surface couples to particle-hole excitations through the Coulomb interaction. This leads to a contribution to the electron-energy-loss spectrum peaked about the specular direction. It is pointed out that if the material is a ferromagnet, a spin dependence of the near-specular loss spectrum occurs as a consequence of interference between the direct Coulomb matrix element, and the non-spin-flip portion of the exchange coupling of the incident electron to the particle-hole spectrum. It is suggested that this interference effect, and not spin-flip scattering, is responsible for the spin-dependent feature reported by Kirschner, Rebenstorff, and Ibach in their study of the near-specular electron-energy-loss spectrum of the Ni(110) surface. In principle a direct experimental test of this proposal is possible.

I. INTRODUCTION

Electron-energy-loss spectroscopy has evolved into an important technique for the study of elementary excitations on and near-crystal surfaces.[1] The analysis of the vibrational normal modes of adsorbed species and of atoms in the crystal surface are a primary application of the technique, but electronic excitations may be studied as well. In the near-specular scattering geometry, the long-ranged direct Coulomb interaction between the incoming electron and those in the sample allow electron-hole excitations (and collective modes) to produce a feature with loss cross section peaked near the specular direction,[2–5] reminiscent of dipole scattering from vibrational modes.

Recently, it has proved possible to produce spin-polarized electron beams sufficiently monoenergetic to allow study of loss spectra, and spin-dependent effects in these features, from ferromagnetic samples. The spin of the scattered electron may also be detected; this allows explicit study of the spin-flip contribution to the loss spectrum. An example is provided by the recent work of Kirschner, who studied off-specular losses from electron-hole excitations in Fe, with both the spin of the incident and scattered electron resolved.[6]

In this paper, we wish to discuss aspects of the particle-hole contribution to the electron-energy-loss spectrum in the near-specular regime. The discussion is motivated by earlier data reported by Kirschner, Rebenstorff, and Ibach.[7] These authors explored the loss spectrum for scattering of a spin-polarized beam off the Ni(110) surface; only data taken on the specular was reported, and the spin of the scattered electron was not resolved. The authors see a spin dependence in the spectrum, with a larger loss cross section when the incident electron spin is antiparallel to the magnetization of the sample. The spin-dependent contribution to the cross section (after averaging over a range of incident energies) is a broad feature, peaked at 300 meV.

Following Yin and Tosatti,[8] Kirschner *et al.*[7] argue that spin-flip processes with origin in exchange scattering are responsible for the spin dependence of the loss feature. The spectrum then provides a measure of the exchange splittings in the d bands near the X point of the Brillouin zone of ferromagnetic Ni; the wave-vector dependence of the exchange splitting was assumed responsible for the very substantial width of the loss structure.

In this paper, we suggest that an alternate mechanism may lead a feature such as that reported by Kirschner and collaborators. Consideration of spin-rotation invariance requires the presence of a non-spin-flip contribution to the exchange scattering amplitude, in addition to the spin-flip portion. There is then interference in the loss cross section between the scattering amplitude produced by this term, and the direct Coulomb coupling of the incident electron to the non-spin-flip electron-hole excitations. As we shall see, this produces a spin-dependent feature in the data that is expected to be distributed over a broad region of energy loss. We point out that through a suitable experiment in which the spin of the scattered electron is resolved, with spin-polarized beam incident, one may discriminate clearly between the mechanism discussed in Refs. 7 and 8, and that explored here.

The outline of this paper is as follows. In Sec. II, we discuss the reasons we seek an alternate explanation of the data, and introduce the process explored here. In Sec. III we present a model calculation which illustrates some key features of the interference mechanism, and general discussion is in Sec. IV. Concluding remarks appear in Sec. V.

Before we turn to the topic of this paper, we note that an interference effect similar to that explored here provides the dominant source of longitudinal magnetoresistance of dilute magnetic alloys.[9] Also, in an early discussion of exchange scattering of electrons from magnetic surfaces, Vredevoe and de Wames[10] recognized the presence of the non-spin-flip portion of the exchange scattering amplitude, and explored implications of interference between it and the direct Coulomb term in the elastic scattering from magnetic surfaces studied in low-energy electron diffraction (LEED) studies of ferromagnets.

II. GENERAL DISCUSSION

The peak in the energy-loss data reported by Kirschner *et al.* occurs at 300 meV, after averaging over a range of incident beam energies, as remarked in Sec. I. This is a value close to the exchange splitting of the Ni *d* bands, as inferred from spin-polarized photoemission data.[11]

However, it is not obvious how other features of the data may be accounted for, on the basis of the exchange scattering mechanism proposed by Yin and Tosatti.[8] In Ref. 7, it is pointed out that the intensity of the entire loss spectrum, including the spin-dependent portion, decreases markedly as one moves off the specular direction. Kirschner comments that the decrease in intensity is several orders of magnitude.[12]

It is well established that the direct Coulomb coupling between the incoming electron, and (non-spin-flip) particle-hole excitations leads to such behavior in the loss cross section.[1] The particle-hole excitations produce fluctuations $\delta\rho(\mathbf{x},t)$ in the charge density of the substrate, and these charge fluctuations generate long-ranged fluctuating electric fields in the vacuum above the crystal. Such long-ranged fluctuating fields lead to a strong peak in the loss cross section at small-momentum transfer,[13] a phenomenon familiar from other physical situations where a charged particle scatters from long-ranged fields.

An exchange process, which involves an electron many volts above the vacuum and a second one in an occupied state in the substrate, necessarily involves a matrix element of the Coulomb interaction evaluated at large-momentum transfer. The dependence on the momentum transfer suffered by the beam electron is thus very weak when small-angle, near-specular scatterings are considered. In these circumstances, quite similar to those of interest to de Wames,[10] the exchange scattering may be viewed as an effective short-ranged interaction, with the coupling constant dependent on beam energy. Spin-flip scattering by such a coupling will not produce a contribution to the loss cross section peaked near the specular direction.

Kirschner *et al.* were fully aware of this point, and suggest[7] that in Ni, the electron energy bands are nearly parallel over an appreciable fraction of the relevant parts of the Brillouin zone. A consequence is that as one moves off the specular direction to scan large momentum transfers, the appropriate loss function will fall off very rapidly with increasing wave-vector transfer. From the viewpoint of the present author, it is unlikely that a special topological property of the energy bands can account for the dramatic decrease reported.

There is not only the spin-flip contribution to the exchange scattering amplitude, but, in addition, there is necessarily a non-spin-flip portion proportional to $s_z S_z$, where s_z is the spin of the beam electron, and S_z that of the electron in the solid which participates in the scattering event. This connects the same initial and final state as the direct Coulomb term, and as a consequence there is an interference term between them in the non-spin-flip channel. As we shall see, this interference term has a sign dependent on the orientation of the spin of the beam electron relative to the host magnetization. We find that its contribution to the cross section is also peaked around the specular direction, because the direct Coulomb matrix element has this property. More precisely, let ψ be the angle between the inelastically scattered electron and the specular direction. The direct Coulomb term has a contribution to the scattering efficiency per unit solid angle which falls off as ψ^{-3}, as one moves off the specular direction. The interference term falls off as ψ^{-2}, the same angular variation found for scattering off the vibrational motions of a monolayer of dipole-active adsorbates.[14]

Kirschner *et al.* also find substantial variations in the shape of the spin-dependent loss feature, as the electron beam energy is varied, and also for small variations in angle of incidence.[15] As we shall see, the interference mechanism allows for such variations, for reasons discussed below. It is a difficult matter to reproduce the variations found in the experiment, with the simple model that forms the basis of the present discussion.

If the mechanism explored here is indeed responsible for the spin-dependent energy loss explored on the specular direction by Kirschner *et al.*, then the peak in the energy-averaged loss spectrum is unrelated to the Stoner splitting of the *d* bands. Examination of the complex band structures proposed for Ni near the *X* point of the Brillouin zone shows many allowed transitions in the range of 100—500 meV, so the energy averaged spectrum is a complex average over these. Agreement of the peak with the Stoner splitting is then fortuitous if the mechanism explored here is dominant, though clearly the largest contributions to the loss cross section will come from this energy range.

It is difficult to envision a fully quantitative analysis of the interference effect explored in this paper for a realistic model of Ni. Fortunately, there is a clear experimental test of the proposal that the interference term is responsible for the loss features reported in Refs. 7 and 15. As remarked earlier, the experiments on Ni(110) employed a spin-polarized primary beam, but the spin of the scattered electron was not resolved. If the spin is resolved, and the interference term rather than spin-flip scattering is responsible for the near-specular, spin-dependent losses, then there should be a near-specular peak only in the two non-spin-flip channels; there should be only a modest angular variation in the spin-flip channel and, in fact, the spin-flip intensities should remain rather weak on specular, as they are off specular. Thus, the Ni(110) experiment should be repeated, if possible, but with spin-polarized primary beam, and the spin of the scattered electron resolved. While Kirschner has explored electron energy loss from Fe surfaces with spin of both incident and scattered electron resolved,[6] he unfortunately reports no data taken on the specular.

The mechanism of Yin and Tosatti provides a natural explanation for the *sign* of the asymmetry in the cross section. The interference term can have either sign, in principle. In fact it is possible, again in principle, for the sign of the asymmetry to change as the beam energy is varied. The reader shall appreciate the fact that it will prove difficult to predict the sign unambiguously from theory.

III. A MODEL CALCULATION

When the direct Coulomb coupling to particle-hole excitations is explored theoretically, a simple and schematic description of the elastic scattering of the electron from the substrate proves fully adequate[1,2,13] for quantitative purposes. The reason, as remarked in Sec. I, is that the charge fluctuations $\delta\rho(\mathbf{x},t)$ produce a long-ranged Coulomb field seen by the electron as it approaches or exits from the crystal; the electrons suffer small-angle inelastic scattering when they are quite far above the crystal, and the role of the elastic scattering from the substrate is simply to turn the electron around, to cause it to reemerge near the specular (or a Bragg) direction. This may be described by appending the appropriate elastic scattering amplitude, calculated from LEED theory, to the description of the inelastic event.[13]

Exchange scattering is described by an effective interaction of short range,[10] and a consequence is that the inelastic events occur after the electron enters the crystal, and while it is engaged in the sequence of multiple (elastic) scatterings that turn it around, to reemerge into the vacuum. A fully microscopic description which incorporates the loss event with multiple scattering (elastic) from the substrate atom cores is required for a proper description of such processes. Such a theory would be similar to that which has been developed and implemented to describe the scattering of electrons from surface phonons in the off-specular (impact) regime, where long-ranged fields are unimportant.[16] The exchange matrix element, an object considerably more complex than the electron-phonon matrix element employed in the theory of electron—surface-phonon scatterings, must be constructed and included in the theory.

In view of the complexity of developing a proper theory of exchange coupling to the particle-hole manifold, we shall explore the nature of the interference term of interest in a simple model which employs a phenomenological description of the exchange matrix element, and which does not take explicit account of the multiple scattering of the electron off the atomic cores. From our view, the model is sufficiently complete to provide an outline of the principal qualitative features of the interference term of interest here; these will survive in a fully quantitative theory.

The crystal is viewed as semi-infinite in extent, residing in the lower half space $z<0$. The charge fluctuations $\delta\rho(\mathbf{x},t)$ produced by the particle-hole excitations generate a potential[13]

$$\phi_1(\mathbf{x},t)=e\int_{z'<0}\frac{d^3x'\,\delta\rho(\mathbf{x}'t)}{|\mathbf{x}-\mathbf{x}'|} \tag{3.1}$$

seen by the electron as it approaches or exits from the crystal in the vacuum $z>0$, and while the electron is in the crystal it experiences the exchange coupling

$$\phi_2(\mathbf{x},t)=\Theta(-z)V_cJ\mathbf{s}\cdot\mathbf{S}(x,t)\ , \tag{3.2}$$

where $\mathbf{s}$ is the spin of the beam electron, $\mathbf{S}(\mathbf{x},t)$ the spin density of the electrons in the substrate, and V_c is the volume of the unit cell, inserted so J has the units of energy, and $\Theta(x)=+1$ when $x\geq 0$, and $\Theta(x)=0$ when $x<0$.

We also include the inner potential $V_0(z)=V_0\Theta(-z)$, where V_0 is complex, the imaginary part describing the attenuation of the electron beam as it enters the crystal. We should take explicit account of the presence of the ion cores, possibly in a muffin-tin description, but this complication we set aside for the moment.

Following an approach outlined earlier,[13] we analyze the Schrödinger equation of the beam electron, in units with $\hbar=1$,

$$\left[-\frac{\nabla^2}{2m}+V_0(z)+\phi(\mathbf{x},t)\right]\psi(\mathbf{x},t)=i\frac{\partial}{\partial t}\psi(\mathbf{x},t)\ . \tag{3.3}$$

Here, $\phi(\mathbf{x},t)=\phi_1(\mathbf{x},t)+\phi_2(\mathbf{x},t)$, and we shall treat the influence of this term in the Born approximation.

As in earlier treatments,[13] it is useful to perform a partial Fourier transform of the wave function, with the subscript $\|$ denoting the projection of a vector onto a plane parallel to the surface:

$$\psi(\mathbf{x},t)=\int\frac{d^2k_\| d\omega}{(2\pi)^3}\psi(\mathbf{k}_\|\omega;z)e^{i\mathbf{k}_\|\cdot\mathbf{x}_\|}e^{-i\omega t}\ . \tag{3.4}$$

We also write

$$\delta\rho(\mathbf{x},t)=\int\frac{d^2Q_\| d\Omega}{(2\pi)^3}\rho(\mathbf{Q}_\|\Omega;z)e^{i\mathbf{Q}_\|\cdot\mathbf{x}_\|}e^{-i\Omega t}\ , \tag{3.5}$$

with a similar relation for the spin density $\mathbf{S}(\mathbf{x},t)$. Then after some algebra, the Schrödinger equation is transformed to read

$$\left[\frac{k_\|^2}{2m}-\omega-\frac{1}{2m}\frac{d^2}{dz^2}+V_0(z)\right]\psi(\mathbf{k}_\|\omega;z)$$
$$=\int\frac{d^2Q_\| d\Omega}{(2\pi)^3}\Phi(\mathbf{Q}_\|\Omega;z)\psi(\mathbf{k}_\|-\mathbf{Q}_\|,\omega-\Omega;z)\ , \tag{3.6a}$$

where

$$\Phi(\mathbf{Q}_\|\Omega;z)=\frac{2\pi e}{Q_\|}\int_{-\infty}^{0+}dz'e^{-Q_\||z-z'|}\rho(\mathbf{Q}_\|\Omega;z)$$
$$+\Theta(-z)V_cJ\mathbf{s}\cdot\mathbf{S}(\mathbf{Q}_\|\Omega;z)\ . \tag{3.6b}$$

We proceed, as earlier, by introducing a Green's function which satisfies

$$\left[\frac{k_\|^2}{2m}-\omega-\frac{1}{2m}\frac{d^2}{dz^2}+V_0(z)\right]G(k_\|\omega;zz')=\delta(z-z')\ , \tag{3.7}$$

and boundary conditions appropriate to the scattering problem. Then, if $\psi_0(\mathbf{k}_\|\omega;z)$ is a solution of Eq. (3.6a) with $\Phi=0$, arranged to describe an electron incident on the surface, we have

$$\psi(\mathbf{k}_\|\omega;z)=\psi_0(\mathbf{k}_\|\omega;z)+\int\frac{d^2Q_\| d\Omega}{(2\pi)^3}\int_{-\infty}^{+\infty}dz'G(\mathbf{k}_\|\omega;zz')\Phi(\mathbf{Q}_\|\Omega;z')\psi(\mathbf{k}_\|-\mathbf{Q}_\|,\omega-\Omega;z')\ . \tag{3.8}$$

A Born-approximation description of the (weak) inelastic scattering is generated by inserting $\psi_0(\mathbf{k}_{\|}\omega;z)$ in the second term on the right-hand side of Eq. (3.8).

The function $\psi_0(\mathbf{k}_{\|}\omega;z)$ describes an electron with a given spin-orientation incident on the crystal. We assume the electron spin is either parallel or antiparallel to the bulk magnetization. Then we shall explore here only scatterings in which the spin of the scattered electron is *parallel* to that of the incident electron, i.e, we examine non-spin-flip processes.

We label the direction of the spin of the incoming electron with an index σ. If $\sigma=+1$, the beam electron has spin parallel to the bulk magnetization, and when $\sigma=-1$, it is antiparallel. If $S_z(\mathbf{Q}_{\|}\Omega;z)$ is the operator which describes spin fluctuations parallel to the bulk magnetization, we define

$$\Phi(\mathbf{Q}_{\|}\Omega;z)=\frac{2\pi e}{Q_{\|}}\int_{-\infty}^{0+}dz' e^{-Q_{\||}|z-z'|}\rho(\mathbf{Q}_{\|}\Omega;z') + \tfrac{1}{2}V_c J\sigma\Theta(-z)S_z(\mathbf{Q}_{\|}\Omega;z)\ . \quad (3.9)$$

The scattered wave, $\psi_s(\mathbf{k}_{\|}\omega;z)$ is then

$$\psi_s(\mathbf{k}_{\|}\omega;z)=\int\frac{d^2Q_{\|}d\Omega}{(2\pi)^3}\int_{-\infty}^{+\infty}dz' G(\mathbf{k}_{\|}\omega;zz')\Phi_{\|}(\mathbf{Q}_{\|}\Omega;z')\psi_0(\mathbf{k}_{\|}-\mathbf{Q}_{\|},\omega-\Omega;z')\ . \quad (3.10)$$

In our mixed representation, the incident wave to be inserted into Eq. (3.10) has the form[13]

$$\psi_0(\mathbf{k}_{\|}\omega;z)=(2\pi)^3\delta(\mathbf{k}_{\|}-\mathbf{k}_{\|}^{(I)})\delta(\omega-E^{(I)})\begin{cases}[e^{-ik_\perp^{(I)}z}+R_>^{(I)}e^{ik_\perp^{(I)}z}], & z>0\\ T_>^{(I)}e^{-iK_\perp^{(I)}z}, & z<0\ .\end{cases} \quad (3.11)$$

In Eq. (3.10), $E^{(I)}$ is the energy of the incident electron, and $\mathbf{k}_{\|}^{(I)}$ the projection of its wave vector on a plane parallel to the surface. Then

$$k_\perp^{(I)}=[2mE^{(I)}-(k_{\|}^{(I)})^2]^{1/2}$$

is the component of the electron's wave vector normal to the surface, in the vacuum above the surface, and

$$K_\perp^{(I)}=[2m(E^{(I)}-V_0)-(k_{\|}^{(I)})^2]^{1/2}$$

is the (complex) wave vector of the incident electron, normal to the surface, when it is in the crystal. We always choose $K_\perp^{(I)}$ so that $\mathrm{Im}(K_\perp^{(I)})>0$. Finally, $R_>^{(I)}$ is the reflection coefficient of the electron off the surface and $T_>^{(I)}$ that for the transmission through it, when the electron is incident on the surface from above.

The form of the Green's function is found in earlier papers.[13,14] Here we only require its form when $z>0$, since we need the scattered wave function as $z\to\infty$. We have, with $k_\perp$ and $K_\perp$ the wave-vector components normal to the surface for an electron of energy ω and wave-vector projection $\mathbf{K}_{\|}$ onto the surface plane,

$$G(\mathbf{k}_{\|}\omega;zz')=i\frac{m}{k_\perp}e^{ik_\perp z}\left\{\Theta(z')[e^{-ik_\perp z'}+R_>e^{+ik_\perp z'}]+\Theta(-z')\frac{k_\perp}{K_\perp}T_<e^{-iK_\perp z'}\right\}\ . \quad (3.12)$$

In Eq. (3.10), $T_<$ is the amplitude which describes transmission of the electron through the surface, when it is incident from below, within the crystal.

When Eq. (3.11) and Eq. (3.12) are inserted into Eq. (3.10), the integrations on z' may be carried out, while we may also let $z\to\infty$. If we append the superscript s to the variables which refer to the scattered electron, invoke the set of approximations used earlier[13] to evaluate the small-angle scatterings produced by the long-ranged Coulomb field, and define

$$\Delta k_\perp=k_\perp^{(s)}-k_\perp^{(I)}\ , \quad (3.13a)$$

$$\bar{K}_\perp=\tfrac{1}{2}(K_\perp^{(s)}+K_\perp^{(I)})\ , \quad (3.13b)$$

$$\bar{R}_>=\tfrac{1}{2}(R_>^{(s)}+R_>^{(I)})\ , \quad (3.13c)$$

then we have for the amplitude of the scattered wave in the non-spin-flip channel

$$\psi_\sigma^{(s)}(k_{\|}\omega;z)=ie^{ik_\perp^{(s)}z}\left[\frac{4\pi me\bar{R}_>}{Q_{\|}^2+(\Delta k_\perp)^2}\int_{-\infty}^{0+}dz'' e^{Q_{\|}z''}\rho(Q_{\|}\Omega;z'')+\frac{\sigma JV_c}{2}\frac{mT_>^{(I)}T_<^{(s)}}{K_\perp^{(s)}}\int_{-\infty}^{0}dz'' e^{-2i\bar{K}_\perp z''}S_z(\mathbf{Q}_{\|}\Omega;z'')\right]\ , \quad (3.14)$$

where $T_<$ describes the amplitude for transmitting an electron wave through the surface, when it is incident from below.

We may now use the amplitude of the scattered wave in Eq. (3.14) to form an expression for the scattering efficiency per unit solid angle, per unit energy loss, i.e., we form the quantity $[d^3S/d^2\Omega(\hat{k}_s)d\omega]$, where $[d^3S/d^2\Omega(\hat{k}_s)d\omega]d^2\Omega(\hat{k}_s)d\omega$ is the probability the electron is scattering into the solid angle $d^2\Omega(\hat{k}_s)$, with energy loss in the range $\hbar$ to $\hbar\omega+\hbar d\omega$ ($\hbar$ added).

We express the result in terms of certain correlation functions, defined as follows. Let $A(\mathbf{x},t)$ and $B(\mathbf{x},t)$ be the Heisenberg operators which represent two dynamical variables. To be explicit, we write these as $A(\mathbf{x}_{\|}z,t)$ and $B(\mathbf{x}_{\|}z,t)$. Then we introduce the correlation function

$$\Gamma_{AB}(\mathbf{Q}_{\|}\Omega;z'z)=\int d^2x_{\|}dt e^{i\mathbf{Q}_{\|}\cdot\mathbf{X}_{\|}} \times e^{-i\Omega t}\langle A(\mathbf{x}_{\|}z',t)B(0z;0)\rangle_T , \quad (3.15)$$

where $\langle\cdots\rangle_T$ denotes the ensemble average of the enclosed operators, over the statistical ensemble at temperature T.

To simplify the answer, we overlook the small difference in the incident and scattered electron energies, in quantities that vary slowly with this variable, i.e., a product like $T_{>}^{(I)}T_{<}^{(s)}$ is replaced by simply $T_{>}T_{<}$. Then after some calculation, we find

$$\frac{d^3S}{d^2\Omega(\hat{k}_s)d\omega}=\frac{m^2(k^{(I)})^2\cos\theta_I}{(2\pi)^3}\times\left[\frac{16\pi^2e^2|R_{>}|^2}{k_{\perp}^2[Q_{\|}^2+(\Delta k_{\perp})^2]^2}\int_{-\infty}^{0+}dz\int_{-\infty}^{0+}dz' e^{Q_{\|}(z+z')}\Gamma_{\rho\rho}(\mathbf{Q}_{\|}\Omega;z'z)\right.$$
$$+\frac{2\pi e\sigma JV_c}{k_{\perp}[Q_{\|}^2+(\Delta k_{\perp})^2]}\left\{\frac{R_{>}T_{>}^{*}T_{<}^{*}}{K_{\perp}^{*}}\int_{-\infty}^{0}dz\int_{-\infty}^{0}dz' e^{i2K_{\perp}^{*}z'}e^{Q_{\|}z}\Gamma_{S_z\rho}(\mathbf{Q}_{\|}\Omega;z'z)+\frac{R_{>}^{*}T_{>}T_{<}}{K_{\perp}}\times\int_{-\infty}^{0}dz\int_{-\infty}^{0}dz' e^{-i2K_{\perp}z}e^{Q_{\|}z'}\Gamma_{\rho S_z}(\mathbf{Q}_{\|}\Omega;z'z)\right\}$$
$$\left.+\frac{J^2V_c^2|T_{>}|^2|T_{<}|^2}{4|K_{\perp}|^2}\int_{-\infty}^{0}dz\int_{-\infty}^{0}dz' e^{-i2K_{\perp}z'}e^{+i2K_{\perp}z}\Gamma_{S_zS_z}(\mathbf{Q}_{\|}\Omega;z'z)\right] \quad (3.16)$$

The result in Eq. (3.16) is the central result of the present paper, expressed in its most general form. We write this as

$$\frac{d^3S}{d^2\Omega(\hat{k}_s)d\omega}=\frac{d^2S^{(a)}}{d^2\Omega(\hat{k}_s)d\omega}+\sigma\frac{d^2S^{(b)}}{d^2\Omega(\hat{k}_s)d\Omega}+\frac{d^2S^{(c)}}{d^2\Omega(\hat{k}_s)d\omega} , \quad (3.17)$$

to emphasize that the second and third terms in Eq. (3.16) have a sign dependent on the orientation of the spin of the beam electron relative to the substrate magnetization. These have their physical origin in the interference between the direct (long-ranged) Coulomb coupling to the density fluctuations produced by particle-hole excitations, and exchange coupling between the electron and the *longitudinal* fluctuations in spin density (to which spin flips do not contribute).

The first term in Eq. (3.16), involving $\Gamma_{\rho\rho}$ is precisely the contribution explored earlier.[2,13] For scattering near the specular direction, they may be related to the long-wavelength, complex dielectric constant $\epsilon(\Omega)$ of the substrate. One finds

$$\frac{d^3S}{d^2\Omega(k_s)d\omega}=\frac{4e^2m^2Q_{\|}|R_{>}|^2n(\Omega)}{\pi^2\cos(\theta_I)[Q_{\|}^2+(\Delta k_{\perp})^2]^2}\times\mathrm{Im}\left[\frac{-1}{1+\epsilon(\Omega)}\right] , \quad (3.18)$$

where $n(\Omega)=[\exp(\hbar\Omega/k_BT)-1]^{-1}$ is the Bose-Einstein function.

Let ψ be the angle between the scattered electron beam direction, and the specular direction, with $\psi_E=\hbar\Omega/2E^{(I)}$, and let $k^{(I)}$ be the wave vector of the incoming electron. Then one finds that[14]

$$Q_{\|}^2+(\Delta k_{\perp})^2=(k^{(I)})^2(\psi^2+\psi_E^2) . \quad (3.19)$$

As one moves away from the specular direction ($\psi>>\psi_E$), $Q_{\|}\sim\psi$, so the cross section displayed in Eq. (3.18) falls off as ψ^{-3}.

The fourth term in Eq. (3.16) describes the inelastic scattering from longitudinal spin fluctuations, produced by the short-ranged exchange coupling. All quantities which enter this expression vary smoothly, as one scans through the specular direction, so there is no tendency for these to be a prominent peak in the intensity of this portion of the loss cross section near specular, such as that which we see in Eq. (3.18). We shall not explore the properties of this term further.

We shall not discuss scattering by spin-flip processes explicitly in this paper, but will comment briefly on their contribution to the loss cross section. Spin-flip scattering is produced by the s_+S_- and s_-S_+ terms in the exchange interaction. If, in the present model, we analyze these, of course there is no interference with the density fluctuation scatterings. The contribution to the cross section by spin-flip processes is given by an expression very similar to the fourth term in Eq. (3.16), but $\Gamma_{S_zS_z}$ is replaced by the correlation function $\Gamma_{S_+S_-}$, which describes the transverse fluctuations in spin density.

In our model, the study of near specular losses probes the response function at large momentum transfer, since scattering off the spin fluctuations must "turn the electron around," so it reemerges near the specular direction. A full description, including multiple scattering off the ion cores allows the sequence of elastic scatterings experienced by the electron redirect it to specular; it may then emerge near the specular by a small-momentum-transfer inelastic event. Such events were assumed dominant in the discussion presented in Refs. 7 and 8. But small-angle, spin-flip inelastic scatterings probe the response function $\Gamma_{S_+S_-}$ at small wave vectors, and here a collective description of transverse spin fluctuations must necessarily be employed. The use of the single-particle picture, such as that invoked in Ref. 8, is qualitatively incorrect here. In the long-wavelength regime, spin waves very nearly saturate the sum rule on the spectral density, and the Stoner excitations make a very small contribution as a consequence.[17] This has been known for many years,[18] and in the view of the present author raises additional difficulties with the interpretation of the kind of data offered in Refs. 7 and 8.

We now turn to the second and third terms in Eq. (3.16), which are the primary focus of the present discussion.

The prefactor in the spin-dependent terms falls off more slowly far off the specular than that in $d^3S^{(a)}/d^2\Omega(\hat{k}_s)d\omega$; we see a ψ^{-2} variation, rather than the ψ^{-3} dependence discussed above. The former is in fact identical (when $\psi \gg \psi_E$) to the angular variation displayed by the loss cross section for scattering off of a dipole active monolayer of vibrating adsorbates. Quite clearly, from the experimental point of view, the prefactor in $d^2S^{(b)}/d\Omega(\hat{k}_s)d\omega$ falls off rapidly enough for the spin-dependent loss feature from the interference term to be sharply peaked near the specular direction.

We now turn to further study of the interference term. We shall be concerned with its general structure here, then turn to model descriptions in Sec. III.

From the general structure of the correlation function in Eq. (3.15), one may show that

$$\Gamma_{S_z\rho}(\mathbf{Q}_{\parallel}\Omega;zz')=[\Gamma_{\rho S_z}(\mathbf{Q}_{\parallel}\Omega;z'z)]^* . \tag{3.20}$$

Upon collecting prefactors together, we have

$$\frac{d^3S^{(b)}}{d^2\Omega(k_s)d\omega}=\frac{em^3JV_cv^{(I)}}{2\pi^2[Q_{\parallel}^2+(\Delta k_{\perp})^2]}\text{Re}\left[\frac{R_>^*T_>T_<}{K_{\perp}}\int_{-\infty}^{0}dz\,dz'e^{Q_{\parallel}z'}e^{-i2K_{\perp}z}\Gamma_{\rho S_z}(\mathbf{Q}_{\parallel}\Omega;z'z)\right] . \tag{3.21}$$

Here, $v^{(I)}$ is the velocity of the incoming electron.

We shall wish to explore the properties of $\Gamma_{\rho S_z}(\mathbf{Q}_{\parallel}\Omega;z'z)$. First, consider the Fourier transform $\Gamma_{AB}(\Omega)$ of the correlation function $\langle A(t)B(0)\rangle$. We have

$$\Gamma_{AB}(\Omega)=\int_{-\infty}^{+\infty}dt\,e^{+i\Omega t}\langle A(t)B(0)\rangle . \tag{3.22}$$

It is well known that we may relate $\Gamma_{AB}(\Omega)$ to the Fourier transform $\chi_{AB}(\Omega)$ of the susceptibility

$$\chi_{AB}(t)=-i\Theta(t)\langle[A(t),B(0)]\rangle , \tag{3.23}$$

where

$$\chi_{AB}(t)=\int\frac{d\Omega}{2\pi}e^{-i\Omega t}\chi_{AB}(\Omega) . \tag{3.24}$$

One finds, in the present notation, with $n(\Omega)$ the Bose-Einstein function encountered earlier,

$$\Gamma_{AB}(\Omega)=\frac{1}{i}n(\Omega)[\chi_{AB}(\Omega+i\eta)-\chi_{AB}(\Omega-i\eta)] . \tag{3.25}$$

The physical interpretation of $\chi_{AB}(t)$ is as follows. Suppose we add to the system Hamiltonian the term $\lambda A\exp(-i\Omega t)+\text{H.c.}$ (in the Schrödinger representation), then calculate the expectation value of the operator B at time $t=0$, after the perturbation is switched on adiabatically from time $t=-\infty$. Then, in essence, $\lambda\chi_{AB}(\Omega)$ is equal to $\langle B\rangle$ at $t=0$.

We then see that $\Gamma_{\rho S_z}$ is found by considering the *density fluctuation* induced by an oscillatory *magnetic field* applied parallel to the z axis, parallel to the magnetization of the sample.

If we consider a paramagnetic, itinerant electron gas in zero dc magnetic field, then $\Gamma_{\rho S_z}$ vanishes identically. In essence, the up- and down-spin electrons reside in identical energy bands; the up-spin electrons respond exactly 180° out of phase with respect to the down-spin electrons (in linear response theory), and no density fluctuation is induced by such a magnetic field.

However, if we consider a spin-polarized, itinerant electron gas, then by virtue of the exchange splitting, the up- and down-spin Fermi surfaces differ, the response of the up- and down-spin electrons differ in magnitude and possibly in phase. A consequence is that application of a longitudinal magnetic field induces a fluctuation in density. Low[19] presented the first discussion of static disturbances in both charge and spin density produced by an external perturbation applied to a spin polarized electron gas, and a more complete discussion appears in the subsequent work of Cullen,[20] and of Kim *et al.*[21]

Thus, the interference term vanishes for scattering off a paramagnetic substrate as it must, of course, since there can be no dependence of the loss cross section on the incident spin direction. The reader should keep in mind that the fourth term in Eq. (3.16), denoted as $d^3S^{(c)}/d^2\Omega(\hat{k}_s)d\omega$ in Eq. (3.17), is nonzero even for the paramagnetic case. The spin-flip amplitude is also nonzero, for scattering from a paramagnetic gas, quite clearly.

In the discussions of Refs. 7 and 8, it is assumed that since the exchange scattering takes place while the electron is inside the material, and at the beam energies used (5—20 eV), the electron penetrates several atomic layers, the loss cross section probes bulk properties of the material. We follow by replacing $\Gamma_{\rho S_z}(\mathbf{Q}_{\parallel}\Omega;zz')$ by a bulk form that depends on only the difference $(z-z')$. Noting the relation in Eq. (3.24), we write with $\mathbf{Q}=\mathbf{Q}_{\parallel}+\hat{z}q_{\perp}$,

$$\Gamma_{\rho S_z}(\mathbf{Q}_{\parallel}\Omega;z'z)=\int_{-\infty}^{+\infty}\frac{dq_\perp}{2\pi}e^{iq_\perp(z-z')}S(\mathbf{Q},\Omega)=\frac{1}{i}n(\Omega)\int_{-\infty}^{+\infty}\frac{dq_\perp}{2\pi}e^{iq_\perp(z-z')}[\chi_{\rho S_z}(\mathbf{Q},\Omega+i\eta)-\chi_{\rho S_z}(\mathbf{Q},\Omega-i\eta)]\ . \quad (3.26)$$

The integrals on z and z' which remain in Eq. (3.21) may then be carried out, to give, after writing $K_\perp=K_\perp^{(1)}+iK_\perp^{(2)}$,

$$\frac{d^3S^{(b)}}{d^2\Omega(\hat{k}_s)d\omega}=\frac{em^3JV_c v^{(I)}n(\Omega)}{2\pi^2[Q_{\parallel}^2+(\Delta k_\perp)^2]}\mathrm{Re}\left[\frac{R_>^*T_>T_<}{iK_\perp}\int_{-\infty}^{+\infty}\frac{dq_\perp}{2\pi}\frac{[q_\perp^2+iQ_{\parallel}(2K_\perp^{(1)}-iK_\perp^{(2)})]}{[q_\perp^2+Q_{\parallel}^2][(q_\perp-2K_\perp^{(1)})^2+(K_\perp^{(2)})^2]}\times[\chi_{\rho S_z}(\mathbf{Q},\Omega+i\eta)-\chi_{\rho S_z}(\mathbf{Q},\Omega-i\eta)]\right] \quad (3.27)$$

To cast Eq. (3.27) in the form stated, we have assumed $\chi_{\rho S_z}(\mathbf{Q},\Omega\pm i\eta)$ is an even function of $q_\perp$, for fixed $\mathbf{Q}_{\parallel}$, an assumption valid under the conditions of interest.

Under typical experimental conditions, where near-specular loss spectra are explored, we expect $Q_{\parallel}\cong 10^6$ cm^{-1}, while $K_\perp^{(1)}\cong 10^8$ cm^{-1} and $K_\perp^{(2)}\cong 10^7$ cm^{-1}. For these estimates, recall that l, the depth of penetration of the electron beam into the material, is $l=(K_\perp^{(2)})^{-1}$. A study of the integrand under these conditions shows that the region of small $q_\perp$, where $q_\perp\cong Q_{\parallel}$, makes a modest contribution. The dominant piece comes from the region $q_\perp\cong 2K_\perp^{(1)}$ where, by virtue of the fact that $K_\perp^{(2)}\ll K_\perp^{(1)}$, the integrand is peaked. We may ignore the factor $iQ_{\parallel}(2K_\perp^{(1)}-iK_\perp^{(2)})$ in the numerator, and replace $q_\perp^2/(q_\perp^2+Q_{\parallel}^2)$ by unity with little error. Then

$$\frac{d^3S^{(b)}}{d^2\Omega(\hat{k}_s)d\omega}=\frac{em^3JV_c v^{(I)}n(\Omega)}{2\pi^2[Q_{\parallel}^2+(\Delta k_\perp)^2]}\mathrm{Re}\left[\frac{R_>^*T_>T_<}{iK_\perp}\int_{-\infty}^{+\infty}\frac{dq_\perp}{2\pi}\frac{[\chi_{\rho S_z}(q_\perp,\Omega+i\eta)-\chi_{\rho S_z}(q_\perp,\Omega-i\eta)]}{(q_\perp-2K_\perp^{(1)})^2+(K_\perp^{(2)})^2}\right]\ . \quad (3.28)$$

In $\mathbf{Q}=\mathbf{Q}+\hat{z}q_\perp$, we also ignore the small quantity $\mathbf{Q}_{\parallel}$.

At large wave vectors, we expect $\chi_{\rho S_z}(q_\perp,\Omega\pm\eta)$ to vary slowly with $q_\perp$, over a region whose width is $K_\perp^{(2)}$. We thus replace $q_\perp$ by $2K_\perp^{(1)}$ here, and then the integral on $q_\perp$ is elementary. With $l=(K_\perp^{(2)})^{-1}$ the penetration depth of the electron beam into the substrate, we have

$$\frac{d^3S^{(b)}}{d^2\Omega(\hat{k}_s)d\omega}=\frac{em^3lJV_c v^{(I)}n(\Omega)}{(2\pi)^2\hbar^5[Q_{\parallel}^2+(\Delta k_\perp)^2]}\times\mathrm{Re}\left[\frac{R_>^*T_>T_<}{iK_\perp}[\chi_{\rho S_z}(2K_\perp^{(1)},\Omega+i\eta)-\chi_{\rho S_z}(2K_\perp^{(1)},\Omega-i\eta)]\right]\ . \quad (3.29)$$

The remainder of this paper will be based on Eq. (3.29). The reader may be disturbed to see a single factor of the electron charge e in the prefactor. Keep in mind that here, ρ is the electron charge density, so the correlation function $\chi_{\rho S_z}$ also contains a factor e. The scattering efficiency is then proportional to e^2, as expected. We have also inserted $\hbar$ explicitly into Eq. (3.29).

We discuss the consequences of Eq. (3.29) in Sec. IV.

IV. GENERAL DISCUSSION

The response function in Eq. (3.29) is evaluated at large-momentum transfer, $2K_\perp^{(1)}$, which is typically a large wave vector, the order of that at the Brillouin-zone boundary. It is then evident that Eq. (3.29) describes a large-momentum-transfer inelastic loss. After the electron enters the crystals, it is the inelastic event, not elastic scattering from the ion cores which "turns it around," so it reemerges near the specular direction.

Of course, a proper theory which takes due account of multiple scattering from the ion cores will include multiple elastic scatterings which can turn the electron around. Near specular, electron-loss spectroscopy generally probes inelastic events in which the momentum transfer experienced by the electron is very small. In the discussion of Ref. 7, it was assumed that such small-momentum-transfer inelastic events were being probed, and then to understand the large width observed in the energy averaged loss spectra, it was necessary for the authors to invoke the presence of a large wave-vector dependence in the exchange splitting in the d bands of ferromagnetic Ni.

We shall argue below that, at least in regard to the loss mechanism explored here, the contribution from large-momentum-transfer inelastic events will dominate that from small-momentum-transfer inelastic scatterings, assisted by elastic scattering that turns the electron around. Our crude model describes the former adequately, in our opinion.

To continue, we require the form of the correlation function $\chi_{\rho S_z}(q,\Omega\pm i\eta)$. Before we begin, we note that in addition to the papers mentioned earlier,[19–21] which explore only simple models, Callaway and co-workers have discussed the structure of such response functions for real metals,[22] although his primary attention is directed to the study of spin-wave excitations. We begin with a general discussion of its structure, for the case where we have simple bands of noninteracting electrons, spin polarized as in an itinerant ferromagnet. We can suppose we are describing the system in the density functional formalism, and after the wave functions and effective energy bands are obtained, we calculate the dynamic response function as if the ensemble of electrons experience nothing other than the crystal potential, supplemented by the appropriate static (spin-dependent) exchange and correlation potential. This will allow us to appreciate the general structure of the correlation function, and we turn to further consequences of the electron-electron interactions later.

Such electrons are described by a field operator

$$\psi(\mathbf{x},t)=\sum_{\mathbf{k},\sigma,j}\phi_{\mathbf{k}j\sigma}(\mathbf{x})C_{\mathbf{k}j\sigma}\exp[-iE_\sigma(\mathbf{k}j)t] \ , \qquad (4.1)$$

where $\phi_{\mathbf{k}j\sigma}(\mathbf{x})$ is the relevant Bloch function, $E_\sigma(\mathbf{k}j)$ is its energy, while the wave vector $\mathbf{k}$ lies within the first Brillouin zone, j is a band index, and σ is a spin index. We assume ϕ normalized so $\int d^3x\,|\phi|^2=1$. In this discussion, we ignore spin-orbit coupling, so $\phi_{\mathbf{k}j\sigma}(\mathbf{x})$ is an eigenfunction of S_z, with eigenvalue $\sigma/2$, where σ is ±1.

Then $\chi_{\rho S_z}(\mathbf{q},\Omega-i\eta)$ is defined as follows, when the substrate is viewed as a structureless entity as in the discussion of Sec. III:

$$\chi_{\rho S_z}(q,\Omega-i\eta)=-i\int d^3x\int_0^\infty dt e^{i\mathbf{q}\cdot(\mathbf{x}'-\mathbf{x})}e^{-i(\Omega-i\eta)t}\langle[\rho(\mathbf{x},t),S_z(\mathbf{x}',0)]\rangle \ . \qquad (4.2)$$

If we employ real Bloch functions, as in Eq. (4.1), then in our description of the energy-loss event, we are implicitly recognizing structure in the substrate. The discussion in Sec. III may be extended to this case (with the elastic scattering off the substrate treated as before), and the result is that $\chi_{\rho S_z}(q,\Omega-i\eta)$ is replaced by

$$\chi_{\rho S_z}(q,\Omega-i\eta)=-i\int\frac{d^3x d^3x'}{V}\int_0^\infty dt\, e^{i\mathbf{q}\cdot(\mathbf{x}-\mathbf{x}')}e^{i(\Omega-i\eta)t}\langle[\rho(\mathbf{x},t),S_z(\mathbf{x}',0)]\rangle \ , \qquad (4.3)$$

a result which reduces to Eq. (4.2) when structure is ignored in the substrate, and the correlation function depends on only $(\mathbf{x}-\mathbf{x}')$. In Eq. (4.3), V is the volume of the substrate. With

$$\rho(\mathbf{x},t)=e\sum_{\mathbf{k},\mathbf{q}}\sum_{j,j'}\sum_{\sigma}\phi^*_{\mathbf{k}j\sigma}(\mathbf{x})\phi_{\mathbf{k}+\mathbf{q},j',\sigma}C_{\mathbf{k}j\sigma}C_{\mathbf{k}+\mathbf{q},j',\sigma}\exp\{i[E_\sigma(\mathbf{k}j)-E_{\sigma'}(\mathbf{k}+\mathbf{q},j')]t\} \ , \qquad (4.4)$$

and a similar expression for $S_z(\mathbf{x}',0)$, we have[23]

$$\chi^{(0)}_{\rho S_z}(q,\Omega\pm i\eta)\frac{e}{2V}\sum_{\mathbf{k}}\sum_{j,j'}|\langle\mathbf{k}j|e^{-i\mathbf{q}\cdot\mathbf{x}}|\mathbf{k}+\mathbf{q},j'\rangle|^2\sum_\sigma\sigma\frac{[f_\sigma(\mathbf{k}j)-f_\sigma(\mathbf{k}+\mathbf{q},j')]}{E_\sigma(\mathbf{k}j)-E_\sigma(\mathbf{k}+\mathbf{q},j')-Q\mp i\eta} \qquad (4.5)$$

The superscript zero appended to $\chi_{\rho S_z}$ in Eq. (4.5) emphasizes the fact that we have ignored the role of electron-electron interactions so far, with the exception of their role in influencing the form of the crystal potential, through the exchange correlation potential. We define the spectral density function $A^{(0)}_{\rho S_z}(\mathbf{q},\Omega)$ through the relation, with N the number of unit cells in the crystal,

$$\begin{aligned}eA^{(0)}_{\rho S_z}(\mathbf{q},\Omega)&=\frac{1}{i}V_c[\chi^{(0)}_{\rho S_z}(\mathbf{q},\Omega+i\eta)-\chi^{(0)}_{\rho S_z}(\mathbf{q},\Omega-i\eta)]\\&=\frac{\pi e}{N}\sum_{\mathbf{k}}\sum_{j,j'}|\langle\mathbf{k}j|e^{-i\mathbf{q}\cdot\mathbf{x}}|\mathbf{k}+\mathbf{q},j'\rangle|^2\sum_\sigma[f_\sigma(\mathbf{k}j)-f_\sigma(\mathbf{k}+\mathbf{q},j')]\delta(E_\sigma(\mathbf{k}j)-E_\sigma(\mathbf{k}+\mathbf{q},j')-\hbar\Omega) \ . \end{aligned}\qquad (4.6)$$

Upon noting that $A^{(0)}_{\rho S_z}(\mathbf{q},\Omega)$ is real, Eq. (3.29) becomes

$$\frac{d^3S^{(b)}}{d^2\Omega(\hat{k}_s)d\omega}=\frac{e^2m^3lJv^{(I)}n(\Omega)}{(2\pi)^2\hbar^4[Q_{\parallel}^2+(\Delta k_\perp)^2]}\times\mathrm{Re}\left[\frac{R^*_>T_>T_<}{K_\perp}\right]A^{(0)}_{\rho S_z}(2K_\perp^{(1)},\Omega) \ . \qquad (4.7)$$

There are both intra- and interband contributions to the loss cross section. Because the spectral function which appears in Eq. (4.7) is evaluated at large-momentum transfer, little can be said in general about the shape of the loss cross section, without a rather detailed calculation. However, it is clear that both the scattering from intra- and interband transitions will produce the sort of broad loss structure evident in the data. Furthermore, in Ni, the Fermi energy is sufficiently close to the top of the d band, and the bands are sufficiently close together that the primary features will lie in the 100—500 meV range.

Suppose we wish to invoke the present mechanism, within a picture that supposes that elastic scattering experienced by the electron when it is inside the material turns it around, so the loss process is a necessarily small angle, small-momentum-transfer event. Then in Eq. (4.6), the wave vector $\mathbf{q}$ will be quite small ($\sim10^6$ cm^{-1}), and the matrix element which controls the strength of the interband processes will be very small. As $\mathbf{q}\to0$, the matrix element vanishes by orthogonality. Indeed, since the matrix element vanishes as $\mathbf{q}\to0$, the leading contribution to it will be proportional to components of $\mathbf{q}$, and the relevant contribution to the loss cross section will no longer be peaked about the specular. For the case where the inelastic loss turns the electron around, the matrix element remains insensitive to angles near the specular, and we retain the peak. Thus, if we wish to invoke the mechanism explored here, insofar as interband processes are concerned, it is the process contained within our simple model that is of interest.

We next turn to the contribution from intra-band scatterings. The simplest case to consider is that of a simple band of electrons, with wave-vector-independent exchange splitting. If we use plane wave eigenstates, then

$$A^{(0)}_{\rho S_z}(\mathbf{q},\Omega)=\frac{\pi}{N}\sum_{\mathbf{k}}\{[f_\uparrow(\mathbf{k})-f_\uparrow(\mathbf{k}+\mathbf{q})]-[f_\downarrow(\mathbf{k})-f_\downarrow(\mathbf{k}+\mathbf{q})]\}\times\delta(E(\mathbf{k})-E(\mathbf{k}+\mathbf{q})-\Omega)\ . \quad (4.8)$$

The case of Ni is modeled by supposing that the majority of the up-spin band is filled, so for $A^{(0)}_{\rho S_z}(\mathbf{q},\Omega)$ we have

$$A^{(0)}_{\rho S_z}(\mathbf{q},\Omega)=\frac{\pi}{N}\sum_{\mathbf{k}}[f_\downarrow(\mathbf{k}+\mathbf{q})-f_\downarrow(\mathbf{k})]\times\delta(E(\mathbf{k})-E(\mathbf{k}+\mathbf{q})-\Omega)\ , \quad (4.9)$$

which is a well-known function, the imaginary part of the particle-hole propagator. We again have a broad loss feature in the large-momentum-transfer regime distributed over energies characteristic of the band structure.

Quite clearly, electron-electron interaction effects will, and in some cases, in a dramatic manner, modify conclusions based on the free-particle description of contributions to the response.

One may appreciate this by noting that, as discussed in Sec. III, $\chi_{\rho S_z}(q,\Omega)$ measures the amplitude of the disturbance in electron density, produced by a time- and space-varying magnetic field parallel to the magnetization. The resulting density fluctuation will then be screened by the electrons themselves, with the result that, particularly at small wave vectors, the amplitude of the density fluctuation will be heavily modified.

One may explore this effect, within a random-phase-approximation (RPA) description of a single band of electrons which interact via the Coulomb interactions. We omit a detailed description of the calculation, which is straightforward. With exchange ignored, the result obtained is precisely that expected from the remarks of the preceding paragraph:

$$\chi_{\rho S_z}(q,\Omega)=\frac{1}{\epsilon(q,\Omega)}\chi^{(0)}_{\rho S_z}(q,\Omega)\ , \quad (4.10)$$

where $\epsilon(q,\Omega)$ is the frequency and wave-vector-dependent dielectric constant.

At the large-momentum transfer $2K^{(1)}_{\perp}$ which enters Eq. (3.29), the screening from the factor of $\epsilon(q,\Omega)$ is of modest importance, but if small-momentum-transfer processes are explored, $\epsilon(q,\Omega)$ may be replaced by $\epsilon(0,\Omega)$ to very good approximation. In the infrared frequency range relevant to the experiments which motivate this work, for Ni, $\epsilon(0,\Omega)$ decreases rapidly with increasing frequency, with both real and imaginary parts of comparable magnitude.[24] It is difficult to make a definitive statement about the contribution of small-angle loss processes to the cross section in the presence of screening, without detailed knowledge of $\chi^{(0)}_{\rho S_z}(q,\Omega)$ for the appropriate band structure. (Once again, such processes require multiple scattering inside the substrate to return the electron to the specular direction, so a small-momentum-transfer loss suffices.) We have attempted to model the contribution of small-angle losses to the cross section with the parabolic band model, to encounter difficulty producing a structure similar to the data, when the factor of $\epsilon(0,\Omega)$ is included. Since $\epsilon(0,\Omega)$ decreases in magnitude with increasing Ω in the infrared, with the d holes modeled by a parabolic band, the loss feature increases with Ω, with no tendency to fall off. This, combined with the fact that for $q\cong 0$, screening reduces the magnitude of $A_{\rho S_z}$, suggests that small-angle intra-band inelastic processes are unimportant.

It is possible to extend the RPA description of $\chi_{\rho S_z}(q,\Omega)$ to a real band structure, in a calculation that incorporates screening in combination with both inter and intra-band processes. Such a calculation is beyond the scope of this paper, and would prove most useful. What is observed is a broad loss feature which extends from 100 to 500 meV, and the discussion here suggests that the mechanism under consideration can provide such a feature, though further theoretical study is required before a clear conclusion can be reached on its shape.

V. CONCLUDING REMARKS

The interference effect explored here can account for a number of features in the data reported by Kirschner, Rebenstorff, and Ibach.[7] We have a means of accounting for the peak in the angular variation of the spin-dependent portion of the loss cross section near specular, and it is clear that it will produce a broad loss feature in the correct energy regime, though a quantitative theory of the loss profile requires extensive calculations that go beyond the scope of the present paper.

In the original data, there is a substantial variation in shape of the loss cross section with both beam energy and direction.[15] In fact, at some of the energies used, the measured loss feature shows no sign of falling to zero with increasing loss, for energy losses as large as 500 meV. Such a dependence of the shape of the loss cross section of beam energy and angle is contained in Eq. (3.29). We may expect the complex phase of the product $R^*_> T_> T_<$ to vary with both angle and beam energy while, as explained in footnote (Ref. 20), the difference

$$\frac{1}{i}[\chi_{\rho S_z}(q,\Omega+i\eta)-\chi_{\rho S_z}(q,\Omega-i\eta)]$$

is real only for special, simple models. The complex phase of $R^*_> T_> T_<$ will mix the real and imaginary part of this last quantity in a manner that will vary with energy and angle, to produce a spin-dependent contribution to the loss function which varies in shape with these parameters of the scattering geometry.

Quite clearly, the phase of the product $R^*_> T_> T_<$ plays a key role in controlling the sign of the asymmetry, as one can best appreciate from Eq. (4.7). There is thus no reason to assume the loss cross section is always largest when the spin of the beam electron is aligned antiparallel to the magnetization. Indeed, as a range of energy is scanned, the asymmetry may change sign, at least in principle.

We remind the reader that in Sec. II, we proposed an experimental study which will provide a clear test of the present suggestion, in the absence of a detailed theory of the shape and sign of the expected loss cross section.

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