Spin polarization of secondary electrons in transition metals: Theory

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A theory of the spin polarization of the secondary electrons in transition metals and glasses is presented. In contrast to the secondary-electron intensity distribution, the spin polarization is shown to yield useful information about the electron-electron interaction. The ratio of the lifetimes of majority- to minority-spin electrons can be determined directly from the measured values of the spin polarization $P(E)$ by $\tau_\uparrow(E)/\tau_\downarrow(E)=[(1-p_B)/(1+p_B)][1+P(E)]/[1-P(E)]$, where p_B is the bulk magnetization. The theory is applied to both Fe and Ni.

I. INTRODUCTION

There has been an increasing interest in the spin-dependent properties of magnetic solids over the last few years. An example is the spin polarization of the secondary electrons in magnetic transition metals and related glasses. Secondary electrons are produced by an external beam of monoenergetic electrons incident on the metal. This beam excites electrons in the solid, which in turn excite others, thereby producing a cascade of secondary electrons. Those with energies greater than the work function can escape from the solid and be collected. The secondary-electron-energy distribution, the number of electrons per unit energy as a function of energy has been measured by various workers over a period of many years. It has a roughly similar shape for all materials; it is largest for small energies and falls rapidly with increasing secondary energy. Only in recent years has the spin polarization distribution of the secondaries been measured. The first measurement was carried out by Unguris *et al.*[1] on $Fe_{81.5}B_{14.5}Si_4$ and subsequently measurements were made on Fe,[2] Co,[2] and Ni,[3,4] as well. The shape of the polarization distribution is roughly the same in all cases. At the lowest observable secondary energy, the polarization is two to three times that of the bulk and it falls to the bulk value with increasing secondary energy as indicated in Fig. 1 after Ref. 5. This behavior is only understood in a qualitative way.

It is our purpose to introduce a theory for the secondary-electron polarization. The present paper is an elaboration of an earlier letter.[6] The electron-energy distribution of secondaries is rather insensitive to the details of the electron-electron interaction. Consequently it has been difficult to extract detailed information as to the nature of the scattering. In the case of the polarization distribution, we will see that the experiments are in fact quite sensitive to the form of the electron-electron scattering. In particular, it is possible to extract the ratio of the majority- to minority-spin electron mean-free paths directly from the experimental measurements. Qualitative information can also be obtained regarding the relative magnitude of the exchange matrix element and its energy dependence relative to that for direct scattering.

Our model is based on the following physics: The electron cascade process might be expected to produce secondaries with the same magnetization as the bulk because the vast majority of secondaries are electrons scattered out of the metal ground state rather than redirected primaries. However, in these materials, there is an excess of unfilled minority-spin d states over unfilled majority-spin d states and excited minority-spin electrons can scatter into the former.[7] Minority-spin electrons are therefore scattered out of a given energy at a faster rate than majority-spin electrons. Thus, a net majority polarization is established due solely to the difference in mean free paths. This effect increases at low energies where the scattering into empty d states is emphasized due to the relatively small number of free s-p—like electron states into which scattering can occur. We wish to emphasize that the connection between the spin polarization of the secondaries

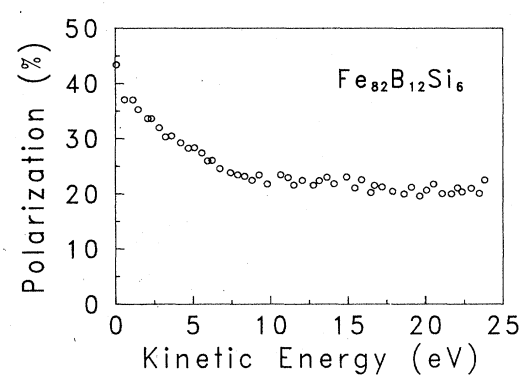

FIG. 1. Plot of the polarization of the secondary electrons versus energy for the ferromagnetic glass $Fe_{82}B_{12}Si_6$. The zero of energy corresponds to the vacuum level. At low energy the polarization is more than twice the bulk polarization of $p_B=22\%$. As the energy increases the secondary-electron polarization rapidly falls to a value close to that of the bulk polarization. This behavior is typical of the magnetic transition metals and glasses; see Figs. 5 and 6 also.

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and the empty d states has been discussed in the experimental papers.[2,3] Our results confirm the importance of this effect. The main result of this paper is given in Eq. (27) which relates the difference in scattering time to the polarization.

The outline of the paper is as follows. In Sec. II, the Wolff model describing the electron cascade process is introduced and generalized to include spin. In Sec. III approximations are introduced for the single scattering probabilities that enter the generalized Wolff theory. In Sec. IV numerical results are presented for the spin polarization distribution and the ratio of the majority to minority spin lifetimes. In Sec. V a simple model for the transition probabilities is introduced. This model allows an analytic solution of the generalized Wolff equation and provides insight into the numerical calculations. Finally, Sec. VI contains the conclusions of the paper.

II. RATE EQUATION FOR THE ELECTRON CASCADE

When a solid is bombarded with electrons of sufficient energy it will emit secondary electrons. The primary electrons penetrate the solid and lose energy creating electron-hole pairs. Those in turn can excite other electron-hole pairs and a cascade of secondary electrons is developed inside the solid. Some of these undergo large-angle scattering, primarily elastic, and can be collected outside the solid in the form of a secondary electron distribution if they have sufficient energy to surmount the surface barrier. In a classic paper, Wolff[8] has developed a formalism using the Boltzmann equation for describing this cascade process. In this section we will repeat some of the basic arguments of Wolff and generalize his theory to include spin.

Inspired by the neutron absorption work of Marshak,[9] Wolff recognized that a Boltzmann transport equation can be used to describe the electron cascade process, via

$$\frac{\partial f}{\partial t}+\frac{1}{m}\mathbf{p}\cdot\nabla f=S-\frac{1}{m}|\mathbf{p}|\frac{f}{\lambda}+\sum_{\mathbf{p}'}f(\mathbf{p}')\omega(\mathbf{p}',\mathbf{p}) , \quad (1)$$

where $f=f(\mathbf{x},\mathbf{p},t)$ is the electron distribution function, S is the source function which describes the rate at which electrons are injected into the solid, $\lambda=\lambda(\mathbf{p})$ the mean free path, and $\omega(\mathbf{p}',\mathbf{p})$ is the probability for an electron to scatter from $\mathbf{p}'$ to $\mathbf{p}$.

In general f is spatially dependent but we will consider the situation in which the penetration of the primaries is such that the production of secondaries takes place uniformly in the bulk over a region which is large compared to that in which surface effects are important. For the energies of interest in the secondary-electron distribution the electron mean-free path is tens of angstroms and neglecting the spatial dependence in f should therefore be a good approximation. This has also been found to be the case in an explicit calculation by Wolff.[8] Moreover we study the situation when we have reached steady state so that $\partial f/\partial t=0$. This gives the following equation for the space- and time-independent secondary-electron distribution $f=f(\mathbf{p})$:

$$f(\mathbf{p})/\tau(E)=S+\sum_{\mathbf{p}'}f(\mathbf{p}')\omega(\mathbf{p}',\mathbf{p}) , \quad (2a)$$

where we have introduced the isotropic lifetime,

$$\frac{1}{\tau(E)}=\frac{|\mathbf{p}|}{m}\frac{1}{\lambda(|\mathbf{p}|)} . \quad (2b)$$

Using the semiclassical Boltzmann approach the phase-space cell must be large in comparison to a lattice cell. We thus ignore the effects of local fields on λ and have therefore replaced $\lambda(\mathbf{p})$ by the isotropic form $\lambda(E)$ with $E=\hbar^2p^2/2m$ denoting a free-electron-like state. The transition probability $\omega(\mathbf{p}',\mathbf{p})$ takes explicit account of both electrons scattered "down" to $\mathbf{p}$ from $\mathbf{p}'$ as well as electrons being scattered "up" from the occupied states to $\mathbf{p}$. Equation (2a) thus represents a detailed-balance equation where the rate at which electrons are kicked down to or up to the state $\mathbf{p}$ is balanced by the rate electrons are scattered *out* of the state $\mathbf{p}$ as represented by f/τ. A further simplification of Eq. (2a) comes about by considering the scattering process in detail. Any special angular distribution is soon smeared out by the scattering, especially the elastic large-angle scattering. The net effect is then that the distribution of secondaries at very low energies is almost spherical since these electrons have scattered very many times both inelastically and elastically. Thus to a good approximation we can take the spherical average of Eq. (2a). In doing so we will inevitably discard any contribution from the elastic scattering to the polarization of the secondaries. However, experiment shows this to be a few percent and is thus almost an order of magnitude smaller than the polarization produced in the inelastic collision.

With $\omega(\mathbf{p}',\mathbf{p})$ depending only on the angle between $\mathbf{p}'$ and $\mathbf{p}$ the above discussion leads to the following averaged detailed balance:

$$\bar{f}(E)/\tau(E)=\bar{S}+\int_E^\infty dE'\rho(E')\bar{f}(E')W(E',E) , \quad (3a)$$

where W is the spherical average of ω,

$$W(E',E)=\int\frac{d\Omega}{4\pi}\omega(\mathbf{p}',\mathbf{p}) , \quad (3b)$$

$$\bar{f}(E)=\int\frac{d\Omega}{4\pi}f(\mathbf{p}) , \quad (3c)$$

and $\bar{S}$ is the spherical average of S.

Since we are studying electrons injected high above the Fermi level (E_F) and looking for secondaries a distance in energy of at least the work function above E_F, $\mathbf{p}'$, and $\mathbf{p}$ are assumed to be free-electron-like and $\rho(E)$ is their corresponding density of states. Because $\mathbf{p}$ and $\mathbf{p}'$ are free-electron-like, the average in Eq. (3b) is possible (otherwise it would be difficult to define the angle between them). However, during the scattering process electrons are being excited from occupied to unoccupied bands of different symmetries. This is accounted for by an internal summation in ω which includes a band summation. This will be dealt with below when we write down the explicit form for ω. Multiplying Eq. (3a) by $\rho(E)$ and defining

$$\sum_{\mathbf{p}}f(\mathbf{p})\equiv\int dE\,N(E) , \quad (4a)$$

where

$$N(E)=\bar{f}(E)\rho(E) \quad (4b)$$

is the number of secondaries per unit energy range, we can write

$$\psi(E)=S(E)+\int_E^\infty dE'\,\psi(E')F(E',E)\ , \tag{5a}$$

where

$$\psi(E)\equiv N(E)/\tau(E)\ , \tag{5b}$$

and

$$F(E',E)=W(E',E)\rho(E)\tau(E') \tag{5c}$$

is the probability per unit energy that an electron at E' scatters and an electron is scattered to energy E. This can happen in two ways; the electron at E' can scatter to E or the electron at E' can scatter to some other energy and a ground-state electron is excited to E'. Since our source function is a δ function in energy (the primary beam) the manipulations we have performed in going from Eq. (1) to Eq. (5) only changes its strength, which does not affect the final result since we are interested in quantities such as the polarization where the absolute magnitude of S drops out.

We now include spin, denoted σ ($=\uparrow$ or $\downarrow$) where the majority-spin direction of the sample is taken to be $\uparrow$. Defining $N_\sigma(E)$ as the number of secondary electrons per unit energy interval at energy E with spin σ and $\tau_\sigma(E)$ the lifetime of electrons with spin σ and energy E, Eq. (5) is immediately generalized to

$$\psi_\sigma(E)=S_\sigma(E)+\sum_{\sigma'}\int_E^\infty dE'\,\psi_{\sigma'}(E')F_{\sigma'\sigma}(E',E)\ , \tag{6a}$$

where

$$F_{\sigma'\sigma}=\tau_{\sigma'}(E')\rho_\sigma(E)W(E',\sigma';E,\sigma) \tag{6b}$$

in an obvious notation. It is necessary to sum over all spins σ since F now describes the possibility of spin-flip as well as non-spin-flip processes. For example $F_{\uparrow\downarrow}(E',E)$ is the probability that an electron $E'\uparrow$ scatters and excites a ground-state electron to $E\downarrow$. The spin dependence in the source function reflects the possibility of bombarding the sample with electrons of arbitrary polarization. Measurements are normally carried out in one of two main spin modes. One is to subject the metal to electrons of a particular spin direction and measure the number of electrons coming out of the sample, then to reverse the spin direction and again measure the number of secondaries. Forming the difference between these two populations and dividing by their sum this gives the so-called spin asymmetry. Another method which is the one we will base our calculations on is to have an unpolarized source, that is $S_\downarrow(E)\equiv S_\uparrow(E)$ and then measure the number of spin-up secondaries $N_\uparrow(E)$ and spin-down secondaries $N_\downarrow(E)$ exiting the solid at the particular energy E. This method is also utilized when photon excitation rather than electron excitation is used, but in that case the source function representing the photoelectrons is polarized with the bulk polarization of the sample. In both cases this defines the spin-polarization $P(E)$ of the secondaries at energy E as

$$P(E)=\frac{N_\uparrow(E)-N_\downarrow(E)}{N_\uparrow(E)+N_\downarrow(E)}\ . \tag{7}$$

For later purposes it is convenient to separate out the part of $P(E)$ which depends *directly* on the difference in lifetime between spin-up and spin-down electrons, since as we will show later that this is the key factor responsible for the polarization buildup at low secondary-electron energies. Because of the difference in the available density of states for a spin-up and a spin-down electron in a ferromagnetic material to scatter into, $\tau_\uparrow(E)$ is in general different from $\tau_\downarrow(E)$ especially at low energies where the density of states in the vicinity of the Fermi level for spin-up and spin-down electrons are very different. We thus define

$$\tau^\pm(E)=\tau_\uparrow(E)\pm\tau_\downarrow(E) \tag{8a}$$

and

$$\psi^\pm(E)=\psi_\uparrow(E)\pm\psi_\downarrow(E)\ . \tag{8b}$$

We can then write Eq. (7) as

$$P(E)=\frac{\psi^-/\psi^++\tau^-/\tau^+}{1+(\psi^-/\psi^+)(\tau^-/\tau^+)}\ . \tag{9}$$

Therefore the polarization of the secondaries is given simply by the ratio of lifetimes which is calculated directly from the pertinent transition probabilities and the explicit solution to Eq. (6) which when expressed in terms of ψ^- and ψ^+ becomes

$$\psi^+(E)=S_+(E)+\tfrac{1}{2}\int_E^\infty dE'[\psi^+(F_{\uparrow\uparrow}+F_{\downarrow\downarrow}+F_{\downarrow\uparrow}+F_{\uparrow\downarrow})+\psi^-(F_{\uparrow\uparrow}-F_{\downarrow\downarrow}+F_{\uparrow\downarrow}-F_{\downarrow\uparrow})], \tag{10a}$$

$$\psi^-(E)=S_-(E)+\tfrac{1}{2}\int_E^\infty dE'[\psi^-(F_{\uparrow\uparrow}+F_{\downarrow\downarrow}-F_{\downarrow\uparrow}-F_{\uparrow\downarrow})+\psi^+(F_{\uparrow\uparrow}-F_{\downarrow\downarrow}-F_{\uparrow\downarrow}+F_{\downarrow\uparrow})], \tag{10b}$$

where

$$S_\pm(E)=S_\uparrow(E)\pm S_\downarrow(E)\ . \tag{10c}$$

Notice that $S_-\equiv 0$ for an unpolarized source.

III. MODEL FOR THE SCATTERING PROBABILITIES

In order to solve Eq. (10) we need to specify $F_{\sigma'\sigma}(E',E)$, i.e., to calculate the scattering probability $\omega(\mathbf{p}'\sigma',\mathbf{p}\sigma)$ that an electron in the state $\mathbf{p}'\sigma'$ scatters and produces an electron in the state $\mathbf{p}\sigma$. For nonflip scattering we can write ($\sigma'=\sigma$)

$$\omega(\mathbf{p}',\sigma;\mathbf{p},\sigma)=\frac{2\pi}{\hbar}\sum_{\mathbf{k},\mathbf{k}'}f_\sigma(E_k)[1-f_\sigma(E_{k'})]\,|M^{p'\sigma,k\sigma}_{p\sigma,k'\sigma}-M^{p'\sigma,k\sigma}_{k'\sigma,p\sigma}|^2\delta(E_{p'\sigma}+E_{k\sigma}-E_{k'\sigma}-E_{p\sigma})$$
$$+\frac{2\pi}{h}\sum_{\mathbf{k},\mathbf{k}'}f_{\bar\sigma}(E_k)[1-f_{\bar\sigma}(E_{k'})]\,|M^{p'\sigma,k\bar\sigma}_{p\sigma,k'\bar\sigma}|^2\delta(E_{p'\sigma}+E_{k\bar\sigma}-E_{p\sigma}-E_{k'\bar\sigma})\ , \tag{11a}$$

where $\bar{\sigma}$ is the spin opposite to σ, f is the Fermi distribution factor and $\mathbf{k}$ and $\mathbf{k}'$ are wave vectors in a reduced-zone scheme incorporating a band index. The matrix element is

$$M_{p\sigma,k'\sigma'}^{p'\sigma,k\sigma'} \equiv \langle p'\sigma,k\sigma' \mid V \mid p\sigma,k'\sigma' \rangle \ , \tag{11b}$$

where V is the screened electron-electron interaction. The first term in Eq. (11a) describes the direct and exchange scattering, as shown in Fig. 2(a) and the second term, Fig. 2(b), is a direct term where the electron in the Fermi sea is of opposite spin to the electron in $p'\sigma$.

In the same manner we can write for the scattering probability per unit time for a spin-flip process, from $(\mathbf{p}',\sigma)$ to $(\mathbf{p},\bar{\sigma})$,

$$\omega(\mathbf{p}',\sigma;\mathbf{p},\bar{\sigma}) = \frac{2\pi}{\hbar} \sum_{\mathbf{k},\mathbf{k}'} f(E_{k\bar{\sigma}})[1-f(E_{k'\sigma})] \mid M_{k'\sigma,p\bar{\sigma}}^{p'\sigma,k\bar{\sigma}} \mid^2 \times \delta(E_{p'\sigma}+E_{k\bar{\sigma}}-E_{k'\sigma}-E_{p\bar{\sigma}}) \ , \tag{11c}$$

which is an exchange type term shown in Fig. 2(c). Since we neglect spin-orbit interactions the spin of an electron cannot change; however, it can be "replaced" by an electron of opposite spin from the occupied states as indicated in Fig. 2(c).

Given the transition probabilities we can also calculate the lifetime $\tau_\sigma(E)$. However some care must be exerted since the transition probabilities ω in Eq. (11) depend not only on the incoming electron but on the one being scattered up as well. It is this conversion from one initial electron to two which is the basis for the electron multiplication. Therefore τ is given by

$$\frac{1}{\tau_{\sigma'}(E')} = \tfrac{1}{2} \sum_{\sigma} \int_{E_F}^{E'} dE\, \rho_\sigma(E) W(E',\sigma';E,\sigma) \ , \tag{12}$$

where the factor $\frac{1}{2}$ arises because ω describes two ways of finding an electron at E: it can fall down to E from above or be kicked up to E from below yielding an identical final state. We show in Appendix A that Eq. (12) indeed gives the proper expression for the lifetime. An important constraint comes from Eq. (12). Multiplying it by $\tau_{\sigma'}(E')$ on both sides we identify

$$F_{\sigma'\sigma}(E',E) \equiv W(E',\sigma';E,\sigma)\rho_\sigma(E)\tau_{\sigma'}(E')$$

on the right-hand side, and the following sum rule for the terms entering the kernel in Eq. (10) is obtained:

$$\sum_{\sigma} \int_{E_F}^{E'} dE\, F_{\sigma'\sigma}(E',E) = 2 \ . \tag{13}$$

As recognized by Wolff the sum being two rather than unity accounts for the electron multiplication.

Having thus formulated the necessary ingredients we now proceed to the required transition probabilities. In doing this we make the explicit assumption that the matrix elements invoked are momentum independent. This

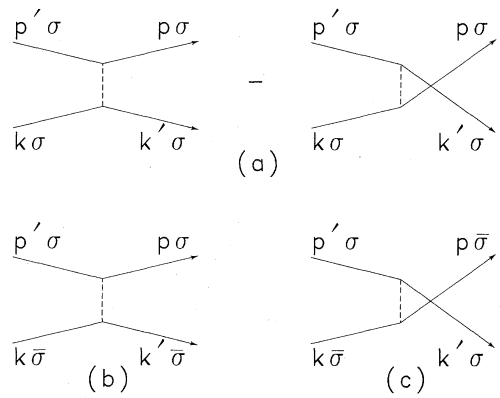

FIG. 2. (a) Direct and exchange scattering of a high-energy electron with momentum p' and spin σ from a ground-state electron $k\sigma$ resulting in electrons in the states (p,σ) and (k',σ). (b) Scattering between opposite spin electrons ($\bar{\sigma}$ is opposite to σ), only direct scattering is possible. (c) Scattering that simulates a spin flip. The ground-state electron $(k,\bar{\sigma})$ is scattered into the state $(p,\bar{\sigma})$.

"random" k approximation has been used easier by Berglund and Spicer[10] and notably by Kane[11] who obtained a very good agreement between the random k approximation and a calculation keeping the full momentum-conservation condition inherent in the matrix elements. This random k approximation moreover has an additional justification when calculations are performed for ferromagnetic glasses which are amorphous in structure and therefore do not give rise to a strict momentum conservation.

In our case this model receives further justification because we perform an explicit angular average, cf. Eq. (3b). In Kane's treatment, the matrix element $M_{p\sigma,k'\sigma'}^{p'\sigma,k\sigma'}$ is replaced by its average value which will be characterized by the energy loss of the primary electron transferred in the collision and the nature of the states $\mathbf{p}$, $\mathbf{p}'$, $\mathbf{k}$, and $\mathbf{k}'$, whether they are free-electron-like (denoted by s) or d-like (denoted by d). For example, $\mid M_{l\sigma,l'\sigma'}^{s\sigma,d\sigma'} \mid^2$ is taken to be $\langle \mid M_{l\sigma,l'\sigma'}^{s\sigma,d\sigma'}(E'-E) \mid^2 \rangle$ if an electron is scattered from $E'(s,\sigma)$ to $E(l,\sigma)$; thus in Eq. (11a) the first matrix elements are replaced by $\langle \mid M(E_{p'}-E_p)-M(E_{p'}-E_{k'}) \mid^2 \rangle$.

The cross terms of this matrix element will be neglected in what follows. This is partly based on the fact that if the momentum dependence is kept in full, the cross term will indeed be small compared to the other terms.[12] For the lifetime where we sum over *all* states $\mathbf{p}$ this is a reasonable approximation, but when calculating $F_{\sigma'\sigma}(E',E)$, especially for low energies, this approximation breaks down and has to be seen as a computational simplification which does not significantly change the outcome of the calculation.

Within the random k approximation, the averaged Eqs. (11a) and (11c) become

$$W(E',\sigma;E,\sigma) = \frac{2\pi}{\hbar} \int d\varepsilon \{ \rho_\sigma^<(\varepsilon) p_\sigma^>(E'-E+\varepsilon)[\mid M(E'-E) \mid^2 + \mid M(E-\varepsilon) \mid^2] + \rho_{\bar{\sigma}}^<(\varepsilon)\rho_{\bar{\sigma}}^>(E'-E+\varepsilon) \mid M(E'-E) \mid^2 \} \tag{14a}$$

and

$$W(E',\sigma;E,\bar{\sigma}) = \frac{2\pi}{\hbar}\int d\varepsilon\,\rho_{\bar{\sigma}}^{<}(\varepsilon)\rho_{\sigma}^{>}(E'-E+\varepsilon)\,|M(E-\varepsilon)|^2 , \tag{14b}$$

where it is to be understood that the M's should be designated with the band labels for the states involved and we have defined

$$\rho_{\sigma}^{<} \equiv f_{\sigma}\rho_{\sigma} \tag{14c}$$

and

$$\rho_{\sigma}^{>} \equiv (1-f_{\sigma})\rho_{\sigma} , \tag{14d}$$

f_{σ} being the Fermi occupation factor which in our calculation is taken to be the zero temperature version, i.e., a sharp Fermi level. ρ_{σ} is the density of states for spin σ electrons.

Because the primary concern is with magnetic properties and because the number of s-p electrons below E_F is small compared to the number of d's, the *occupied* s-p states will not be distinguished from the *occupied* d's whose densities of states will be denoted by $\rho_{\uparrow}^{<}$ and $\rho_{\downarrow}^{<}$ and the total number of spin-up (down) electrons is

$$n_{\uparrow(\downarrow)} = \int_{-\infty}^{E_F} dE\,\rho_{\uparrow(\downarrow)}^{<}(E) , \tag{15}$$

where the integration extends over the occupied s-p and d bands. A characteristic feature of a ferromagnetic material is its bulk polarization p_B given in terms of $n_{\uparrow}$ and $n_{\downarrow}$ as

$$p_B = \frac{n_{\uparrow}-n_{\downarrow}}{n_{\uparrow}+n_{\downarrow}} . \tag{16}$$

It is 6% (17% and 27%) for Ni (Co and Fe). $N \equiv n_{\uparrow}+n_{\downarrow}$ is 10 (9 and 8) for Ni (Co and Fe) and roughly 5% of N comes from the s-p band which justifies not distinguishing it from the d's. An additional simplification is that there are unfilled d states only for minority ($\downarrow$) spin band. This is strictly true for Ni and Co and approximately true for Fe where the majority-spin unfilled d states hold only about 0.2 of an electron. In Fig. 3 we show the density of states as used for Ni in the present calculation indicating our labeling of the occupied and unoccupied states. This density of states is taken from the band-structure calculation Moruzzi *et al.*[13] and for energies greater than those reported in their calculation a free-electron density of states $\rho_0(E)$ is used. Thus in Eq. (14),

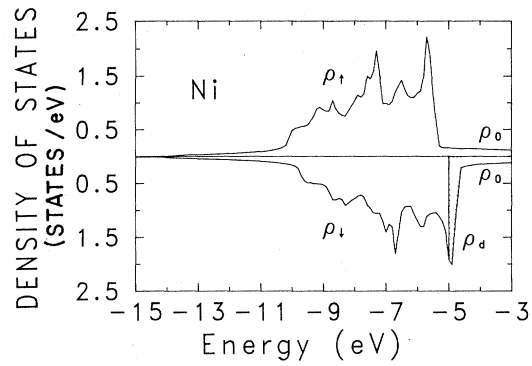

FIG. 3. Densities of majority- (up-) spin and minority- (down-) spin electrons in Ni (states per atom/eV). The Fermi energy is at −5 eV. The occupied states are taken to be d states as are the unoccupied states just above the Fermi energy indicated by the shaded area (ρ_d). The states denoted by ρ_0 are assumed to be free-electron states.

$$\rho_{\uparrow}^{>} = \rho_0(E)[1-f_{\uparrow}(E)] \tag{17a}$$

and

$$\rho_{\downarrow}^{>} = [\rho_0(E)+\rho_d(E)][1-f_{\uparrow}(E)] \equiv \rho_0^{>}+\rho_d^{>} , \tag{17b}$$

where $\rho_d(E)$ is the important *unfilled* minority d-electron density of states. $\rho_d(E)$ is large only in a very small energy range above E_F (shaded area in Fig. 3), and since we will eventually only be interested in the electrons leaving the material we will not consider the scattering which puts the state **p** in the unoccupied d states since these cannot escape. However, these states have to be included when calculating the lifetime.

Having specified the electronic levels involved we now consider in turn the four possible scattering probabilities $F_{\sigma'\sigma}(E',E)$ needed to specify the electron scattering process. Defining

$$A_{\sigma'} \equiv \frac{2\pi}{\hbar}\tau_{\sigma'}(E')\rho_{\sigma}(E) \tag{18}$$

and setting

$$\omega \equiv E'-E$$

and (19a)

$$\omega' \equiv E-\varepsilon ,$$

we can write

$$\frac{F_{\uparrow\uparrow}(E',E)}{A_{\uparrow}} = \int d\varepsilon\{\rho_{\uparrow}^{<}(\varepsilon)\rho_0^{>}(\varepsilon+\omega)[\,|M_{ss}^{sd}(\omega)|^2+|M_{ss}^{sd}(\omega')|^2] + \rho_{\downarrow}^{<}(\varepsilon)[\rho_0^{>}(\varepsilon+\omega)\,|M_{ss}^{sd}(\omega)|^2+\rho_d^{>}(\varepsilon+\omega)\,|M_{sd}^{sd}(\omega)|^2]\} . \tag{19b}$$

The matrix elements are now labeled according to the nature of the states involved in the scattering event.

For E well above E_F ($E-E_F$ at least equal to the work function) we can approximate $\omega' \equiv E-\varepsilon \simeq E-E_F$ and we can therefore take all matrix elements in Eq. (19) outside of the energy integration leaving a joint density of states. Defining

$$\bar{\rho}_{\sigma,l}(\omega) \equiv \int d\varepsilon \rho_\sigma^<(\varepsilon)\rho_l^>(\varepsilon+\omega) , \tag{20}$$

l being s or d, Eq. (19b) can be rewritten yielding

$$\frac{F_{\uparrow\uparrow}(E',E)}{A_\uparrow} \simeq |M_{ss}^{sd}(\omega)|^2[\bar{\rho}_{\uparrow,s}(\omega)+\bar{\rho}_{\downarrow,s}(\omega)] + |M_{ss}^{sd}(E-E_F)|^2\bar{\rho}_{\uparrow,s}(\omega) + |M_{sd}^{sd}(\omega)|^2\bar{\rho}_{\downarrow,d}(\omega) . \tag{21a}$$

Proceeding in the same manner

$$\frac{F_{\downarrow\downarrow}(E',E)}{A_\downarrow} \simeq |M_{ss}^{sd}(\omega)|^2[\bar{\rho}_{\uparrow,s}(\omega)+\bar{\rho}_{\downarrow,s}(\omega)] + |M_{ss}^{sd}(E-E_F)|^2\bar{\rho}_{\downarrow,s}(\omega)$$
$$+ |M_{sd}^{sd}(\omega)|^2\bar{\rho}_{\downarrow,d}(\omega) + |M_{ds}^{sd}(E'-E_F)|^2\bar{\rho}_{\downarrow,d}(\omega) , \tag{21b}$$

$$\frac{F_{\uparrow\downarrow}(E',E)}{A_\uparrow} \simeq |M_{ss}^{sd}(E-E_F)|^2\bar{\rho}_{\downarrow,s}(\omega) , \tag{21c}$$

and

$$\frac{F_{\downarrow\uparrow}(E',E)}{A_\downarrow} \simeq |M_{ss}^{sd}(E-E_F)|^2\bar{\rho}_{\uparrow,s}(\omega) + |M_{ds}^{sd}(E'-E_F)|^2\bar{\rho}_{\uparrow,d}(\omega) . \tag{21d}$$

In Fig. 4 we show diagramatically, in order of appearance in Eq. (21b), the four possible spin-down to spin-down scattering events contributing to F when distinguishing s- and d-electrons. Notice that of all terms contributing to the $F_{\sigma'\sigma}$'s it is only the M_{ds}^{sd} terms of $F_{\downarrow\uparrow}(E',E)$ which leaves the solid with a hole in the spin-up occupied states and an electron in the spin-down unoccupied states, i.e., a Stoner excitation. However, $M_{ds}^{sd} \neq 0$ means that $F_{\uparrow\uparrow}$ differs from $F_{\downarrow\downarrow}$ in general, creating a polarization of the secondaries that does not involve Stoner excitations. We therefore see no justification in recent claims[14] that the Stoner excitation is the dominating source for the low-energy polarization rise in the secondaries; for every event in which a spin-$\downarrow$ electron falls into an empty spin-$\downarrow$ d state and excites a spin-$\uparrow$ electron there is an event in which a spin-$\downarrow$ electron is excited and the net polarization produced cannot be large compared to p_B. In fact it is the empty minority d states which are the common crucial factor, *enabling* Stoner-type excitations and also *other* spin-polarizing scattering events. We will see that it is the difference in $\tau(E_\uparrow)$ and $\tau(E_\downarrow)$ which produces the polarization rise at low energies.

To be able to calculate the distribution of secondaries the lifetimes for spin-up and spin-down electrons are required. From the expression for the lifetime given in Appendix A, Eq. (A1), and the approximation of momentum-independent matrix elements (in reality their angular average) we can write

$$\frac{1}{\tau_\sigma(E')} = \frac{2\pi}{\hbar}\int_{E_F}^{E'} dE\,\rho_\sigma^>(E)\left[\int d\varepsilon'\rho_\sigma^>(\varepsilon')\int d\varepsilon\rho_\sigma^<(\varepsilon)[\tfrac{1}{2}|M(E'-E)|^2+\tfrac{1}{2}|M(E-\varepsilon)|^2]\right.$$
$$\left. + \int d\varepsilon'\rho_{\bar{\sigma}}^>(\varepsilon')\int d\varepsilon\rho_{\bar{\sigma}}^<(\varepsilon)|M(E'-E)|^2\right]\delta(E'+\varepsilon-\varepsilon'-E) , \tag{22}$$

again neglecting the interference term between direct and exchange scattering. Now letting $E \to E'-E+\varepsilon$ in the second term, it becomes equal to the first and we have the much simpler expression

$$\frac{1}{\tau_\sigma(E')} = \frac{2\pi}{\hbar}\int_{E_F}^{E'} dE\,\rho_\sigma^>(E)\left[\int d\varepsilon\rho_\sigma^<(\varepsilon)\rho_\sigma^>(\varepsilon+\omega)+\rho_{\bar{\sigma}}^<(\varepsilon)\rho_{\bar{\sigma}}^>(\varepsilon+\omega)\right]|M(\omega)|^2 . \tag{23}$$

The integrand represents a process in which the spin σ electron at E' is scattered to E and a spin σ or $\bar{\sigma}$ electron in the occupied state ε is excited to $\varepsilon+\omega$. For $\sigma=\uparrow$, we then obtain

$$\frac{1}{\tau_\uparrow(E')} = \frac{2\pi}{\hbar}\int_{E_F}^{E'} dE\,\rho_0^>(E)\int d\varepsilon\{[\rho_\uparrow^<(\varepsilon)+\rho_\downarrow^<(\varepsilon)]\rho_0^>(\varepsilon+\omega)|M_{ss}^{sd}(\omega)|^2+\rho_\downarrow^<(\varepsilon)\rho_d^>(\varepsilon+\omega)|M_{sd}^{sd}(\omega)|^2\} . \tag{24a}$$

In a similar way

$$\frac{1}{\tau_\downarrow(E')} = \frac{2\pi}{\hbar}\int_{E_F}^{E'} dE\,\rho_0^>(E)\left[\int d\varepsilon[\rho_\uparrow^<(\varepsilon)+\rho_\downarrow^<(\varepsilon)]\rho_0^>(\varepsilon+\omega)|M_{ss}^{sd}(\omega)|^2+\rho_\downarrow^<(\varepsilon)\rho_d^>(\varepsilon+\omega)|M_{sd}^{sd}(\omega)|^2\right]$$
$$+\frac{2\pi}{\hbar}\int_{E_F}^{E'} dE\,\rho_d^>(E)\int d\varepsilon[\rho_\uparrow^<(\varepsilon)+\rho_\downarrow^<(\varepsilon)]\rho_0^>(\varepsilon+\omega)|M_{ds}^{sd}(\omega)|^2 . \tag{24b}$$

Notice that it is mainly due to the two last terms in Eq. (24b) which are proportional to M_{ds}^{sd} that $\tau_\downarrow$ is less than $\tau_\uparrow$. Of these two terms one is a Stoner term involving a spin flip in the ground state.

For energies up to about 40 eV we find the energy dependence of the spin-averaged mean free path to be al-

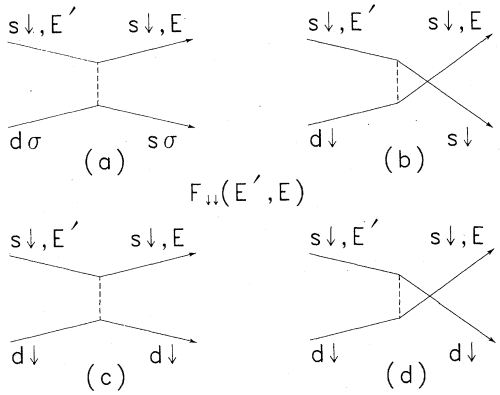

FIG. 4. Terms in Eq. (21b) that contribute to $F_{\downarrow\downarrow}(E',E)$, the probability that a spin ↓ electron with energy E' scatters and an electron is produced with spin ↓ and energy E. (a) The incident electron $s\downarrow,E'$ (where s denotes a free-electron state) scatters from a ground-state d electron of spin σ into the state $(s\downarrow E')$ and the ground state electron is scattered into an empty (s,σ) state. (b) The exchange term corresponding to (a) when σ denotes a spin ↓. (c) Same as (a) except that the ground-state electron $d\downarrow$ is scattered into an empty $d\downarrow$ state rather than an $s\downarrow$ state. (d) Exchange scattering process in which the electron in the state $(s\downarrow,E')$ is scattered into an empty $d\downarrow$ state (near the Fermi energy) and a ground-state electron is scattered to $(s\downarrow,E)$.

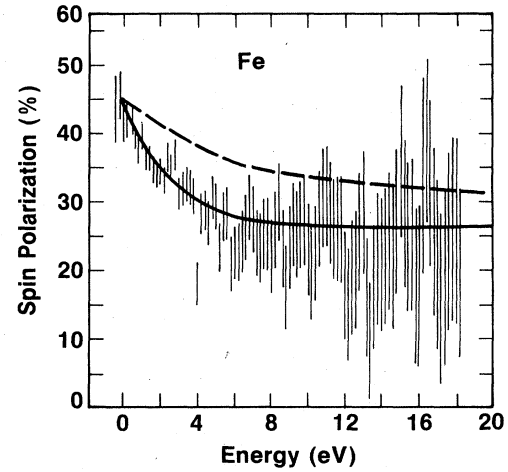

FIG. 5. Polarization of the secondary electrons versus energy in Fe. The experimental results are represented by the vertical lines (Ref. 2). The large error bars are due to the use of photon excitation rather than electron excitation which results in a relatively low number of initially excited electrons. The dashed curve gives the results of the theory using energy-independent matrix elements adjusted to agree with the experimental polarization at zero energy. The solid curve allows for an energy-dependent exchange matrix element, Eq. (26).

most identical to that calculated with the statistical model.[12]

IV. NUMERICAL RESULTS

In this section we discuss numerical results. Combining the above results we can calculate the lifetimes from Eq. (24) and solve Eq. (10) numerically yielding ψ^-/ψ^+. The matrix elements $M_{ss}^{sd}(\omega)$ and $M_{ss}^{sd}(\omega')$ are taken to be energy independent, in which case they are equal and the polarization depends on $|M_{sd}^{sd}(\omega)/M_{ss}^{sd}|^2$ and $|M_{ds}^{sd}(E'-E_F)/M_{ss}^{sd}|^2$. We find empirically that the polarization is very insensitive to the choice of $|M_{sd}^{sd}/M_{ss}^{sd}|^2$ but depends directly on $|M_{ds}^{sd}(E'-E_F)/M_{ss}^{sd}|^2$ as is evident from the spin dependence of Eq. (21). This ratio determines the relative probability that an excited minority-spin electron is scattered into an empty d state. Because of the small effect the size of M_{sd}^{sd} has on the polarization, the somewhat arbitrary choice $|M_{sd}^{sd}/M_{ss}^{sd}|^2=0.3$ in made. The calculations are carried out for a source function that is monoenergetic and polarized with the bulk polarization p_B if photoexcitation has been used (as for Fe) with

$$S_\sigma^{\rm ph}=n_\sigma\delta(E-E_0')\ , \tag{25a}$$

E_0' being the photon frequency. If electron bombardment at energy E_0 is used as in the experiment for Ni, the source function is chosen to be unpolarized, i.e.,

$$S_\sigma^{\rm el}=S_0\delta(E-E_0)\ . \tag{25b}$$

For the case that M_{ds}^{sd} is also energy independent, the choices $|M_{ds}^{sd}/M_{ss}^{sd}|^2=0.13$ for Fe and 0.19 for Ni result in the dashed lines in Figs. 5 and 6. These magnitudes of $|M_{ds}^{sd}/M_{ss}^{sd}|$ are consistent with M_{ds}^{sd} being an exchange-type matrix element. It is apparent from the calculations that the energy dependence of the joint density of states $\bar{\rho}_{\sigma,l}$ is not sufficient to explain the energy dependence of the polarization and the exchange matrix element M_{ds}^{sd} needs to be taken as energy dependent. M_{ds}^{sd} is *extremely* difficult to calculate at low energies because of the importance of screening and correction effects and so we parametrized M_{ds}^{sd} by

$$|M_{ds}^{sd}(E)/M_{ss}^{sd}|^2=\frac{A^2}{(E-E_F)^2+B^2}\ , \tag{26}$$

remembering that over the relevant energy range M_{ds}^{sd} falls off much faster than M_{ss}^{sd}. Recall that $M_{ss}^{sd}=M_{ss}^{sd}(E'-E)$

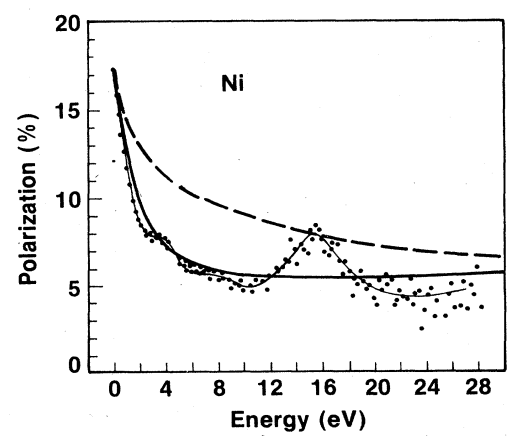

FIG. 6. Polarization of the secondary electrons versus energy in Ni. The experimental results (dots) are from Ref. 3. There is currently no explanation for the peak at 16 eV. The solid and dashed curves have the same meaning as in Fig. 5.

while M_{ds}^{sd} has the argument $E'-E_F$. Since in general $E'-E \ll E'-E_F$, when small energy losses are favored M_{ss}^{sd} is essentially "static" compared with M_{ds}^{sd} which involves a high energy change. Since the matrix elements in general vanish for very large energy transfers Eq. (26) mimics this behavior.

For Fe (Ni) the choice $A=1.8$ eV (2.9 eV) and $B=3.2$ eV (3.5 eV) results in the solid curve shown in Fig. 5 (6). Thus the model we are using with its simplifications, is capable of reproducing the experimental results while retaining as many of the Ni and Fe characteristics as possible and adding no unphysical or unmotivated features.

A more accurate estimate of the ratio of $\tau_\uparrow(E)/\tau_\downarrow(E)$ than the calculated from Eq. (24) can now be obtained from Eq. (9) by using the experimental values of $P(E)$ and the calculated values of $\psi^-(E)/\psi^+(E)$, a quantity that is relatively insensitive to the choice of matrix elements. The quantity $\psi^-(E)/\psi^+(E)$ is found to have a value very close to p_B for values of the parameters discussed above and for $E>0$. It is found empirically and will be justified below that $\psi^-/\psi^+ \simeq p_B$ holds if $|M_{ds}^{sd}/M_{ss}^{sd}|^2 \ll 1$. Replacing ψ^-/ψ^+ in Eq. (9) by p_B we obtain $\tau_\uparrow/\tau_\downarrow$ in terms of the measured polarization

$$\frac{\tau_\uparrow(E)}{\tau_\downarrow(E)}=\frac{1-p_B}{1+p_B}\frac{1+P(E)}{1-P(E)} . \tag{27}$$

Thus, this ratio can be obtained directly from experimental measurements and is largely model independent. This is our central result. The ratio $\tau_\uparrow/\tau_\downarrow$ versus energy is shown in Fig. 7 for Fe and Ni. The ratio is larger in the case of Fe because its ratio of unfilled d states to unfilled free-electron-like states is larger than in the case of Ni, which means that there is an enhanced scattering of minority-spin electrons into empty minority d states.

The results shown in Fig. 7 are in disagreement with previous theories which have neglected the selective scattering of minority spin electrons into empty minority d states. Those that consider exchange only predict[12,15] almost no difference between the up- and down-spin lifetimes while that of Bringer *et al.*[16] assumes $\tau_\uparrow/\tau_\downarrow$

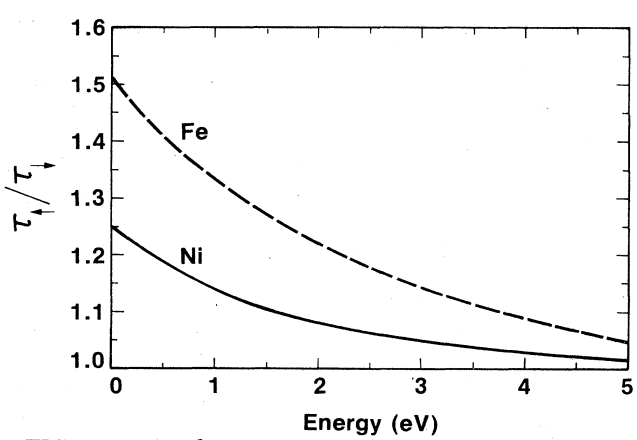

FIG. 7. Ratio of majority- to minority-spin lifetimes for Fe and Ni as determined from Eq. (27) using the experimental polarization data shown in Figs. 5 and 6. The larger ratio for Fe reflects the larger number of unfilled d states in Fe as opposed to Ni.

$=(1+p_B)/(1-p_B)$ independent of energy. We also note that the Stoner excitations observed recently[17,18] in very-low-energy-loss scattering are contained without our theory but are not important to the anomalous secondary-electron polarization because they produce appreciable polarization only for very-small-energy-loss processes.

V. ANALYTIC SOLUTION USING A SIMPLE MODEL FOR SCATTERING PROBABILITIES

We now introduce a simple analytical model to gain insight into the numerical calculations, such as the way in which the polarization depends on the energy of the incoming electrons and the bulk polarization of the sample. We will give explicit results for Ni but the model is easily implemented to treat Fe, Co, and spin glasses as well.

All three of the ferromagnetic metals Fe, Co, and Ni are characterized by a large unfilled minority d-electron peak close to the Fermi level with room for roughly $n_\uparrow - n_\downarrow$ electrons. This peak is replaced with a δ function whose strength is chosen to yield the same area:

$$\rho_d(E) \simeq (n_\uparrow - n_\downarrow)\delta(E-E_d) , \tag{28}$$

where $E_d \simeq E_F$ and $n_\uparrow - n_\downarrow = 0.6$ for Ni.

With constant matrix elements and a free-electron density of states for the unfilled levels which are assumed to be constant $(=\rho_0)$ we obtain the following approximate form for the scattering function $F_{\sigma'\sigma}(E',E)$:

$$\frac{F_{\uparrow\uparrow}(E',E)}{A_\uparrow} \simeq M^2\rho_0(2n_\uparrow+n_\downarrow)+\overline{M}^2(n_\uparrow-n_\downarrow)n_\downarrow\delta(\omega) , \tag{29a}$$

where $A_\uparrow=(2\pi/\hbar)\rho_0\tau_\uparrow(E')$, $\omega=E'-E$, $M=M_{ss}^{sd}$, $\overline{M}=M_{sd}^{sd}$, and we have used

$$\bar{\rho}_{\sigma,s}(\omega)=\int d\varepsilon\,\rho_\sigma^<(\varepsilon)\rho_0^>(\varepsilon+\omega) \simeq \rho_0\int d\varepsilon\,\rho_\sigma^<(\varepsilon)=\rho_0 n_\sigma , \tag{29b}$$

$$\bar{\rho}_{\sigma,d}(\omega)=\rho_\sigma^<(-\omega)(n_\uparrow-n_\downarrow) \simeq n_\sigma\delta(\omega)(n_\uparrow-n_\downarrow) , \tag{29c}$$

where Eq. (29c) is obtained by concentrating all the d states at energy $\omega=0$. In Eq. (29b) we have neglected the fact that for $\omega<E_F$ the integration is not over the whole bandwidth which implies $\bar{\rho}_{\sigma,s}$ is proportional to ω for very small ω. This will influence the lifetime and other quantities very strongly close to the Fermi level but since we deal with energies larger than the work function this has negligible influence on our conclusions.

For $F_{\downarrow\downarrow}(E',E)$ we find

$$\frac{F_{\downarrow\downarrow}(E',E)}{A_\downarrow} \simeq M^2\rho_0(n_\uparrow+2n_\downarrow)+\overline{M}^2(n_\uparrow-n_\downarrow)n_\downarrow\delta(\omega) + \widetilde{M}^2(n_\uparrow-n_\downarrow)n_\downarrow\delta(\omega) , \tag{29d}$$

where $A_\downarrow=(2\pi/\hbar)\rho_0\tau_\downarrow(E')$ and $\widetilde{M}\equiv M_{ds}^{sd}$. Also,

$$\frac{F_{\uparrow\downarrow}(E',E)}{A_\uparrow}=\rho_0 M^2 n_\downarrow \tag{29e}$$

and

$$\frac{F_{\downarrow\uparrow}(E',E)}{A_\downarrow}=\rho_0 M^2 n_\uparrow+\tilde{M}^2(n_\uparrow-n_\downarrow)n_\uparrow\delta(\omega)\ . \tag{29f}$$

The structure of the F's is evident from considering the possible scattering processes. For example, in the case of $F_{\downarrow\uparrow}(E',E)$, the probability that an up-spin electron is excited to E when a down-spin electron scatters is proportional to the number of $\uparrow$-spin electrons present in the occupied states, i.e., $n_\uparrow$. The $\tilde{M}^2$ term describes an electron falling into an empty d state (there are roughly $n_\uparrow-n_\downarrow$ of those) and kicking up an $\uparrow$ electron, i.e., proportional to $n_\uparrow$. Similarly the $\overline{M}$ term is always proportional to $n_\downarrow$ since it corresponds to kicking a $\downarrow$ electron into an empty d state. The δ function in those terms reflects the narrow width of the occupied d states.

Inserting these expressions for the F's into the sum-rule equation, Eq. (13), we find for the lifetimes in the present model (*measuring energies from* E_F)

$$\frac{1}{\tau_\uparrow(E)}=aE+c \tag{30a}$$

and

$$\frac{1}{\tau_\downarrow(E)}=aE+b+c\ , \tag{30b}$$

where we have defined

$$a=\frac{2\pi}{\hbar}NM^2\rho_0^2\ , \tag{31a}$$

$$b=\frac{\pi}{\hbar}N(n_\uparrow-n_\downarrow)\rho_0\tilde{M}^2\ , \tag{31b}$$

and

$$c=\frac{\pi}{\hbar}n_\downarrow(n_\uparrow-n_\downarrow)\rho_0\overline{M}^2\ . \tag{31c}$$

$N\equiv n_\uparrow+n_\downarrow$ and we see that it is $\tilde{M}$ which causes $\tau_\uparrow$ to be different from $\tau_\downarrow$; the scattering process where a spin-down electron falls into the empty minority d states.

The lifetimes in Eq. (30) do not scale as E^2 when E approaches E_F as must be the case from phase space arguments. This is, as pointed out earlier, because of the approximations made in the joint density of states in Eq. (29). However we are only interested in $\tau_\sigma(E)$ for E larger than the work function in which case Eq. (30) introduces a negligible error. When calculating the spin polarization according to Eq. (9) we will only work to lowest order in p_B ($\ll 1$ for Ni). This means

$$P(E)\simeq\psi^-/\psi^++\tau^-/\tau^+\ , \tag{32}$$

because it is evident from Eq. (30) that τ^-/τ^+ is of $O(p_B)$. An inspection of the kernels in Eq. (10), with the F's given by Eq. (29) shows that ψ^- is of $O(p_B)$ since $S_-=0$ for an unpolarized source. Thus it is sufficient to evaluate ψ^+ to lowest order in p_B, i.e., for $n_\uparrow=n_\downarrow$, the paramagnetic solution.

We find from Eq. (30) and the expressions for a, b, and c,

$$\frac{\tau^-}{\tau^+}=p_B\frac{E_1}{E+p_B(E_1+E_2)}\simeq p_B\frac{E_1}{E}+O(p_B^2)\ , \tag{33a}$$

where the characteristic energy scales associated with $\tilde{M}$ and $\overline{M}$ are

$$E_1=\frac{N}{4\rho_0}\frac{\tilde{M}^2}{M^2}\simeq 4E_F\frac{\tilde{M}^2}{M^2}\ , \tag{33b}$$

$$E_2=\frac{N}{4\rho_0}\frac{\overline{M}^2}{M^2}\simeq 4E_F\frac{\overline{M}^2}{M^2}\ , \tag{33c}$$

where $\rho_0 E_F$ is approximately the number of s-p electrons in the filled states, which is $\simeq 0.6$ electrons for Ni.

For the paramagnetic equation for ψ^+ we have, letting $n_\uparrow=n_\downarrow\equiv N/2$ in the F's,

$$\psi^+=S_0\delta(E-E_0)+2\int_E^{E_0}dE'\frac{1}{E'}\psi^+(E')\ . \tag{34}$$

The factor of 2 in front of the integration is the manifestation of the electron multiplication yielding the secondary electron cascade. The solution to Eq. (34) is

$$\psi^+=S_0\delta(E-E_0)+\frac{2S_0E_0}{E^2}\ , \tag{35}$$

which, within the same approximation (paramagnetism) corresponds to a secondary yield $Y\sim\tau\psi^+\sim S_0E_0/E^3$, which increases dramatically at low energies. We also see that we can increase the yield in an obvious fashion by having a stronger source or by letting the incident electron beam have a higher energy E_0.

As mentioned above ψ^- scales with p_B, and working to first order in p_B, defining

$$\psi^-\equiv p_B f(E)\ , \tag{36a}$$

we find the following equation for $f(E)$:

$$f(E)=\int_E^{E_0}dE'\frac{1}{E'}f(E')+\int_E^{E_0}dE'\frac{1}{E'}\left[1+\frac{E_1}{E'}\right]\psi^+(E') \tag{36b}$$

$1/E'$ is the kernel and $f(E)$ has no multiplication factor in front of the integral. $f(E)$ is instead a measure of the spin contrast in the system. Notice that ψ^+ plays the role of a source function for $f(E)$.

The solution to Eq. (36) is easily found to be

$$\psi^-=p_B f(E)=p_B\left[\frac{2S_0E_0}{E^2}-\frac{S_0}{E}+\frac{E_0E_1S_0}{E^3}\right]\ . \tag{37}$$

Thus we obtain for the total polarization, Eq. (32) ($E<E_0$),

$$P(E)=p_B\left[\frac{E_1}{E}+\left[1-\frac{E}{2E_0}+\frac{E_1}{2E}\right]\right]\ , \tag{38}$$

where the first part is the lifetime difference in Eq. (32) and the second comes from solving for ψ^-/ψ^+. There are several notable features of Eq. (38). It scales with p_B; without a bulk polarization, i.e., $n_\uparrow=n_\downarrow$ the secondaries will not be polarized. The lifetime difference dominates for small energies. In fact the $E_1/2E$ in the ψ^-/ψ^+ part is also a direct consequence of $\tau_\uparrow$ being different from $\tau_\downarrow$ entering through $A_{\uparrow(\downarrow)}$ of Eq. (29). It is not the $\overline{M}$ and $\tilde{M}$ terms in F/A which yield this term. Of course, it is these

terms which make $\tau_\uparrow \neq \tau_\downarrow$ when calculating the lifetime but they contribute nothing to $O(p_B)$ in the multiplication equation. Furthermore the $p_B(1-E/2E_0)$ term which arises from ψ^-/ψ^+ is what we would expect from the following consideration: Inject a spin-up and spin-down electron into the solid (unpolarized source), after one scattering event we have four electrons. The two electrons being kicked up should, for large energies, reflect the bulk polarization p_B. In other words, we should start out with a polarization being $(2\times O+2p_B)/4=p_B/2$. However the exchange scattering coming in through $E_1(\tilde{M})$ gives a correction to this $\approx E_1/E_0$, which in general is small since $E_1 \sim 7$ eV for Ni. After the second scattering event there are two unpolarized and six polarized electrons yielding a polarization $(2O+6p_B)/8=p_B(\frac{1}{2}+\frac{1}{4})$, etc. Finally, at very low energies we approach $p_B(\frac{1}{2}+\frac{1}{4}+\frac{1}{8}+\cdots)=p_B$. The exact energy dependence between $P(E_0,E_1=0)=p_B/2$ and $P(O,E_1=0)=p_B$, being linear in this case, can only come from a detailed calculation where the energy dependence of the scattering kernel determines the interpolation between the two limiting results which we have found.

In Fig. 8 we show a calculation of $P(E)$ from Eq. (38) for Ni (solid line), comparing it with the full numerical solution (for constant matrix elements) as elaborated upon earlier in this paper (dashed line). E_1 is calculated from Eq. (33b) using $E_f=9$ eV and $\tilde{M}/M^2=0.19$ in accordance with the full numerical solution. The agreement is very satisfying and we note that the dramatic rise in the secondaries at low energies is evident in the model calculation through E_1 ($\propto \tilde{M}^2/M^2$) and to this order in p_B it is entirely a lifetime difference effect. Out of the two $\tilde{M}$ terms contributing to $F_{\downarrow\downarrow}$ [Eq. (29d)] and $F_{\downarrow\uparrow}$ [Eq. (29f)], it is only the latter one which is a Stoner excitation. Thus of the 18% low-energy secondary-electron polarization $\frac{1}{3}$ is due to the background bulk polarization, $\frac{1}{3}$ is from Stoner excitations and the other $\frac{1}{3}$ is from excitations of spin-down d electrons to spin-down unoccupied d states (making $F_{\uparrow\uparrow}$ and $F_{\downarrow\downarrow}$ different). So we again stress that the necessary condition for having an increased polarization in the secondaries at low energies is to have unfilled minority d states above the Fermi level.

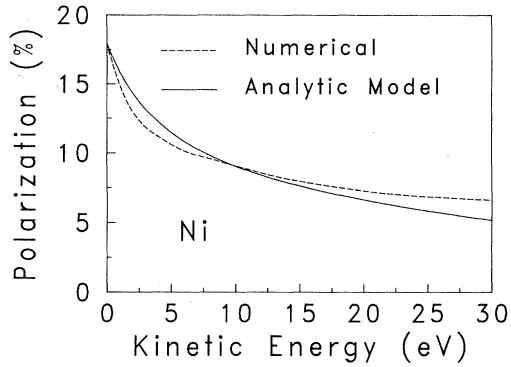

FIG. 8. Comparison of the theoretical results for the polarization of the secondary electrons in Ni assuming constant matrix elements (dashed line) with the results of the simple analytic model, Eq. (38), indicated by the solid line.

We now return to the full Eq. (10) in order to carry out manipulations which shed light on a feature which was first found in the numerical calculations; that apart from energies close to E_0 or below the work function the ratio ψ^-/ψ^+ stays very close to p_B. From Eq. (9) this implies that in this energy interval the polarization increase is mainly due to the difference in lifetime, which grows rapidly as $E\to 0$. We consider Eq. (10) for an unpolarized source, i.e., $S_-=0$ and $S_+=\delta(E-E_0)$. In order to avoid the singularity connected with S_+ we introduce

$$\bar{\psi}^+(E)=\psi^+(E)-S_+(E)\ , \tag{39a}$$

and let

$$\Delta(E)=[\psi^-(E)/\bar{\psi}^+(E)]-p_B\ . \tag{39b}$$

The following equation for $\Delta(E)$ is obtained from Eq. (10b):

$$\Delta(E)=\Big[F(E_0,E)-p_B D(E_0,E)+\int_E^{E_0} dE'\,\bar{\psi}^+(E')(F_{\uparrow\uparrow}+F_{\downarrow\downarrow}-F_{\downarrow\uparrow}-F_{\uparrow\downarrow})\Delta(E') + \int_E^{E_0} dE'\,\bar{\psi}^+(E')[F(E',E)-2p_B(F_{\downarrow\uparrow}+F_{\uparrow\downarrow})]\Big]\Big/2\bar{\psi}^+(E)\ , \tag{40a}$$

where

$$F(E',E)=F_{\uparrow\uparrow}-F_{\downarrow\downarrow}+F_{\downarrow\uparrow}-F_{\uparrow\downarrow}\ , \tag{40b}$$

$$D(E',E)=F_{\uparrow\uparrow}+F_{\downarrow\downarrow}+F_{\downarrow\uparrow}+F_{\uparrow\downarrow}\ , \tag{40c}$$

where it is understood that $F_{\sigma'\sigma}\equiv F_{\sigma'\sigma}(E',E)$ in Eq. (40). $D(E',E)$ is proportional to the total single scattering cross section. The quantity $\bar{\psi}^+(E)$ in Eq. (40) is given by

$$\bar{\psi}^+(E)=\tfrac{1}{2}D(E_0,E)+\tfrac{1}{2}\int_E^{E_0} dE'\,\bar{\psi}^+(E')D(E',E)\ . \tag{41}$$

Equation (41) is derived by neglecting a contribution from ψ^- because $\bar{\psi}^+$ carries the electron multiplication information while ψ^- contains the spin asymmetry [e.g., Eqs. (35) and (37)]. In the sense of a lowest-order calculation, $\bar{\psi}^+$ is only needed to lowest order, i.e., the paramagnetic result, since it is multiplied by $F(E',E)$ in Eq. (40a) which vanishes in this limit because $E_{\uparrow\uparrow}=F_{\downarrow\downarrow}$ and $F_{\uparrow\downarrow}=F_{\downarrow\uparrow}$. Therefore we have neglected the ψ^- contribution to Eq. (41).

We will also introduce another simplification in what follows. We are interested in an energy range between the work function of the material and roughly 10–15 eV above that, which in much less than the typical energies E_0 used for the incoming electrons in the experiments we consider. We thus work in the limit where $E \ll E_0$.

Having introduced $\Delta(E)$, our main objective is to show

that over the pertinent energy range $\Delta(E)$ is indeed small compared to p_B. In fact, this will turn out to be a consequence of the electron multiplication and the condition $M_{ds}^{sd}/M_{ss}^{sd} \ll 1$. In order to focus on the lifetime difference

$$P_\tau \equiv \tau^-/\tau^+ = (\tau_\uparrow - \tau_\downarrow)/(\tau_\uparrow + \tau_\downarrow) , \tag{42}$$

we show in Appendixes B—D how Eq. (40a) can be reduced to

$$\Delta(E) = \Delta_s(E) + \frac{\int_E^{E_0} dE' N(E')(W_{\uparrow\uparrow} - W_{\uparrow\downarrow})\Delta(E')}{\int_E^{E_0} dE' N(E')(W_{\uparrow\uparrow} + W_{\uparrow\downarrow})} , \tag{43a}$$

provided there is an electron cascade and $M_{ds}^{sd} \ll M_{ss}^{sd}$. In Eq. (43a), $N(E)$ is the number of electrons at energy E and $W_{\sigma'\sigma} \equiv W_{\sigma'\sigma}(E',E)$ is given by Eq. (14) and is the probability per unit time that an electron scatters out of the state $E'\sigma'$ and an electron is scattered into $E\sigma$. $\Delta_s(E)$ in Eq. (43a) is given by

$$\Delta_s(E) = \frac{1}{2} \frac{\int_E^{E_0} dE' N(E') P_\tau(E')(W_{\uparrow\uparrow} + W_{\uparrow\downarrow})}{\int_E^{E_0} dE' N(E')(W_{\uparrow\uparrow} - W_{\uparrow\downarrow})} . \tag{43b}$$

We now focus on $\Delta_s(E)$ in order to estimate the size of $\Delta(E)$. We see that the condition for ψ^-/ψ^+ being close to p_B, i.e., $\Delta \ll p_B$ is intimately connected to the average, of the normalized lifetime difference P_τ, as defined in Eq. (42) being small compared to p_B. It should be mentioned here, that in the simple model considered earlier in this section, $N(E) \sim 1/E^3$, $W_{\sigma'\sigma}$ is a constant, and $P_\tau = p_B E_1/E$. This yields $\Delta_s(E) = \frac{1}{3} P_\tau(E)$ which when iterated through the kernel of Eq. (43a) gives $\Delta(E) = \frac{1}{2} P_\tau(E)$ in accord with our previous calculations of Eq. (38) when $E \ll E_0$. In the present model, M_{ds}^{sd} is responsible for the lifetime difference and because the lifetime sum is roughly proportional to M_{ss}^{sd} we see again that in order for $\Delta_s(E)$ to be small M_{ds}^{sd} has to be a small fraction of M_{ss}^{sd}. However the matrix elements are, in general, energy dependent which means that the particular weighting in Eq. (43b) can change the prefactor. In Appendix E this is treated in more detail while we give here a more heuristic plausibility argument that demonstrates $\Delta_s(E) < p_B$ (or P_τ).

It is well known that the secondary-electron cascade goes as $E^{-\beta}$ for low energies. We believe that the $\beta = 3$ obtained from the simple model is a very realistic value. This calculation yielding $\beta = 3$ only depends on the paramagnetic solution and is moreover constrained by the "sum rule" in Eq. (13). For P_τ the situation is less clear (see Appendix E) but we know that P_τ vanishes for high energies and that by construction and the physical processes involved it rises as E decreases to a maximum value of one. We therefore assume P_τ to be of the form

$$P_\tau = p_B (E_\gamma/E)^\gamma , \tag{44}$$

since it has to vanish when $n_\uparrow = n_\downarrow$ ($p_B = 0$). E_γ is a typical energy below which we can expect P_τ to exhibit a saturated behavior (by construction $P_\tau \le 1$). E_γ is intimately connected to M_{ds}^{sd}/M_{ss}^{sd} and since in Eq. (43b) we are integrating from E which is larger than the work function we have a cutoff already built in which means that as long as $E_\gamma \lesssim \Phi$, Eq. (44) is a reasonable estimate. Inserting this into Eq. (43b) we find [assuming $W_{\uparrow\uparrow} + W_{\uparrow\downarrow}$ to be a constant over the range where $N(E')$ varies rapidly]

$$\Delta_s(E) \simeq \frac{1}{2} \frac{\beta - 1}{\beta - 1 + \gamma} P_\tau(E) . \tag{45}$$

Thus, P_τ when it enters the integral Eq. (43) is a algebraically diminished compared to the direct P_τ term entering Eq. (9). Iterating Eq. (43) yields

$$\Delta(E) \simeq \frac{\beta - 1}{\beta - 1 + 2\gamma} P_\tau(E) . \tag{46}$$

This result is consistent with experiment as can be seen from the following arguments. Assume $\Delta(E) \ll P_\tau$. Then we can obtain P_τ directly from the experimental results by subtracting off p_B from the polarization of the secondary electrons as in Eq. (27). For Ni this gives a $\gamma \sim 3$ which together with the reasonable choice $\beta = 3$ gives $\Delta(E) = \frac{1}{4} P_\tau$ which is consistent with our original assumption. We also see that, the faster P_τ increases the smaller Δ is (the larger γ). This means the approximation $\Delta \ll p_B$, i.e., $\psi^-/\psi^+ \simeq p_B$, should be particularly good for Ni where P_τ has to go from $\sim 6\%$ to 100% over an energy interval where iron has to go from $\sim 20\%$ to 100%.

VI. SUMMARY AND CONCLUSIONS

Our theory is based on an equation of detailed balance, Eq. (6), to describe the electron cascade. It simply states that the rate at which electrons are scatterd out of a state $E\sigma$ is equal to the rate at which they are scattered into that state. Equation (6) is a straightforward generalization of a spin-independent theory due to Wolff. It has been assumed that the secondary-electron distribution is isotropic due to elastic scattering. The electron-electron scattering probabilities which enter the theory are extremely difficult to calculate due to correlation and screening effects. The greatest uncertainties are at low energies where even the Born approximation fails. We have parametrized the cross sections by following Kane in assuming momentum-independent matrix elements for the screened Coulomb interactions. This leads to scattering probabilities that are expressed as squares of matrix elements times joint densities of states. The matrix elements are characterized by the nature of the states involved in the collision, free-electron like or d like, and the energy transferred in the collision.

Our principal result is Eq. (27), in which the ratio of the majority- to minority-spin lifetime is related directly to the measured polarization. The inelastic mean free path (or equivalently the lifetime) is a fundamental quantity in the characterization of a solid. A difference between the majority- and minority-spin mean free paths has important implications for the interpretation of many types of spin polarized experiments, including spin-polarized photoemission and inverse photoemission, spin-polarized low-energy electron diffraction, and spin-

polarized elastic scattering from amorphous materials. This is because a difference in mean free paths implies that the polarization P_0 of electrons at a given energy in the bulk of a material differs from that measured outside the material P_M; and this is because in traveling to the surface, more of one spin than the other is scattered out of the beam,

$$P_M=\frac{N_\uparrow\tau_\uparrow-N_\downarrow\tau_\downarrow}{N_\uparrow\tau_\uparrow+N_\downarrow\tau_\downarrow}=\frac{P_0+\tau^-/\tau^+}{1+P_0\tau^-/\tau^+}\ , \tag{47}$$

where $P_0=(N_\uparrow-N_\downarrow)/(N_\uparrow+N_\downarrow)$.

Our results (Fig. 7) show that this effect is important only for very low energies. In this low-energy regime, it has not proved possible to carry out low-energy electron diffraction experiments on Fe and Ni due to stray magnetic fields. The most relevant experiment performed so far is threshold spin polarized photoemission on Ni.[19] As the photon energy is increased from a value equal to the work function, the polarization of the escaping electrons is measured. Electrons scattered to energies below the work function do not escape and the measured polarization is given by Eq. (47). Kisker *et al.*[19] fit their experimental data for Ni (assuming $P_M=P_0$) to a bulk band-structure calculation in which the exchange splitting Δ was taken to be an adjustable parameter and a value of $\Delta=0.33$ eV was obtained. Using Eq. (27) with τ^-/τ^+ determined from Fig. 7 and Ref. 19, we estimate $\Delta\simeq0.37$ eV is required to fit the data. A more interesting case where the correction to the value of Δ would be greater is that of Fe for which τ^-/τ^+ is significantly larger. An analysis similar to that of Ni could provide an independent test of the values of $\tau_\uparrow/\tau_\downarrow$ presented here. Another interesting feature of Fe is that calculations by Victora, Falicov, and Ishida[20] show a significant enhancement of the magnetization at and just below the surface which would affect the spin polarization of the photoemission. It would be useful if the appropriate threshold spin polarized photoemission experiments were carried out in Fe.

There have been two recent experimental papers[17,18] as well as a theoretical paper[21] that studied low energy loss single scattering of electrons from Fe and Ni. All three papers discuss the observed phenomena in terms of Stoner excitations, the scattering of a minority-spin electron and the simultaneous appearance of a majority-spin electron. In general, the Stoner mechanism in which a minority-spin d electron falls into an empty spin-down d state and a spin-up electron is scattered back to high energy is "balanced" by a process in which a spin-down electron is scattered back to high energy. These two processes together can be expected to result only in a spin polarization equal to that of the bulk and are completely taken into account by our theory. The case of very small net energy[17,18,21] loss involves the excitation of electrons very near the Fermi energy and for that case there can be a large imbalance in the number of up- and down-spin electrons and Stoner excitations can dominate.

With the model described above we find:

(1) The rise of the secondary-electron polarization at low energies is due to the scattering of minority-spin electrons into empty minority-spin d states as discussed in Sec. I. This idea has been in the literature for a number of years but has never been employed in a quantitative model of the secondary polarization.

(2) A rough fit to the experimental data can be obtained by assuming energy-independent matrix elements. The exchange-type matrix element that governs the rate at which minority-spin electrons are scattered into empty minority-spin d states M_{ds}^{sd} must be small compared to that describing direct scattering M_{ss}^{sd} in order to fit the data.

(3) To obtain a quantitative fit to the data, the exchange-matrix element must be strongly energy dependent as described by Eq. (26) and must be small compared to the matrix element for direct scattering.

(4) For the case that the exchange-matrix element is small compared to the direct one, as in (2) and (3) above, we find, empirically as well as analytically (in Sec. V), that the measured polarization can be related directly to the ratio of minority to majority electron life times (or equivalently the ratio of mean free paths) which is the main result of the paper [Eq. (27)]. This equation allows the determination of the mean-free-path ratio directly from the experimental data for the polarization without any fitting of matrix elements.

(5) The lifetime difference between minority- and majority-spin electrons is small at energies greater than a few eV above the work function. Thus the experimentally measured polarization is in fact equal to the polarization in the bulk for electrons with energies greater than a few eV.

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APPENDIX A

Consider the lifetime of an electron in state $\mathbf{p}'$ with spin σ, its life time can be written

$$\frac{1}{\tau_\sigma}=\frac{2\pi}{\hbar}\sum_{\mathbf{k},\mathbf{k}',\mathbf{p}}\{f_\sigma(E_k)[1-f_\sigma(E_{k'})][1-f_\sigma(E_p)]\tfrac{1}{2}\,|M_{p\sigma,k'\sigma}^{p'\sigma,k\sigma}-M_{k'\sigma,p\sigma}^{p'\sigma,k\sigma}|^2\delta(E_{p'\sigma}+E_{k\sigma}-E_{p\sigma}-E_{k'\sigma})$$
$$+f_{\bar\sigma}(E_k)[1-f_{\bar\sigma}(E_{k'})][1-f_\sigma(E_p)]\,|M_{p\sigma,k'\bar\sigma}^{p'\sigma,k\bar\sigma}|^2\delta(E_{p'\sigma}+E_{k\bar\sigma}-E_{p\sigma}-E_{k'\bar\sigma})\}\ , \tag{A1}$$

where the factor $\frac{1}{2}$ in the first term of Eq. (A1) is to prevent double counting since the two M's represent the same final state and we are summing over all p. Now in the last term we rewrite $|M_{p\sigma,k'\bar\sigma}^{p'\sigma,k\bar\sigma}|^2$ as $\frac{1}{2}(|\cdots|^2+|\cdots|^2)$ and in one of the terms we exchange $\mathbf{p}$ and $\mathbf{k}'$. This then gives

$$\frac{1}{\tau_\sigma}=\frac{2\pi}{\hbar}\sum_{\mathbf{k},\mathbf{k}',\mathbf{p}}\{f_\sigma(E_k)[1-f_\sigma(E_{k'})][1-f_\sigma(E_p)]\tfrac{1}{2}\,|M^{p'\sigma,k\sigma}_{p\sigma,k'\sigma}-M^{p'\sigma,k\sigma}_{k'\sigma,p\sigma}|^2\delta(E_{p'\sigma}+E_{k\sigma}-E_{p\sigma}-E_{k'\sigma})$$
$$+f_{\bar\sigma}(E_k)[1-f_{\bar\sigma}(E_{k'})][1-f_\sigma(E_p)]\tfrac{1}{2}\,|M^{p'\sigma,k\bar\sigma}_{p\sigma,k'\bar\sigma}|^2\delta(E_{p'\sigma}+E_{k\bar\sigma}-E_{p\sigma}-E_{k'\bar\sigma})\}$$
$$+\frac{2\pi}{\hbar}\sum_{\mathbf{k},\mathbf{k}',\mathbf{p}}f_{\bar\sigma}(E_k)[1-f_\sigma(E_{k'})][1-f_{\bar\sigma}(E_p)]\tfrac{1}{2}\,|M^{p'\sigma,k\bar\sigma}_{k'\sigma,p\bar\sigma}|^2\delta(E_{p'\sigma}+E_{k\bar\sigma}-E_{k'\sigma}-E_{p\bar\sigma})\ , \tag{A2}$$

which we immediately recognize as

$$\frac{1}{\tau_{\sigma'}(E_{p'})}=\sum_{\mathbf{p}}\tfrac{1}{2}\omega(\mathbf{p}',\sigma;\mathbf{p},\sigma)[1-f_\sigma(E_p)]$$
$$+\sum_{\mathbf{p}}\tfrac{1}{2}\omega(\mathbf{p}',\sigma;\mathbf{p},\bar\sigma)[1-f_{\bar\sigma}(E_p)]\ , \tag{A3}$$

with $\omega(\mathbf{p}',\sigma;\mathbf{p},\sigma)$ and $\omega(\mathbf{p}',\sigma;\mathbf{p},\bar\sigma)$ defined in Eqs. (11a) and (11c), respectively. This then immediately leads to

$$\frac{1}{\tau_{\sigma'}(E_{p'})}=\tfrac{1}{2}\sum_{\sigma}\sum_{\mathbf{p}}[1-f_\sigma(E_p)]\omega(\mathbf{p}',\sigma';\mathbf{p},\sigma)\ , \tag{A4}$$

or in terms of our spherical average $W(E',\sigma';E,\sigma)$,

$$\frac{1}{\tau_{\sigma'}(E)}=\tfrac{1}{2}\sum_{\sigma}\int_{E_F}^{E}dE'\rho_\sigma(E')W(E',\sigma';E,\sigma)\ , \tag{A5}$$

which is Eq. (12), hereby proved.

APPENDIX B

In order to focus on the lifetime difference P_τ ($\equiv\tau^-/\tau^+$) in Eq. (42) we explicitly decompose the F's involved according to [cf. Eq. (6b) where $\rho_\uparrow(E)=\rho_\downarrow(E)=\rho_0$ since $E>\phi$, the work function.

$$F(E',E)=\rho_0(E)\frac{\tau^+}{2}(W_{\uparrow\uparrow}-W_{\downarrow\downarrow}+W_{\downarrow\uparrow}-W_{\uparrow\downarrow})$$
$$+\rho_0(E)\frac{\tau^+}{2}P_\tau(W_{\uparrow\uparrow}+W_{\downarrow\downarrow}-W_{\uparrow\downarrow}-W_{\downarrow\uparrow})\ , \tag{B1}$$

$$D(E',E)=\rho_0(E)\frac{\tau^+}{2}(W_{\uparrow\uparrow}+W_{\downarrow\downarrow}+W_{\uparrow\downarrow}+W_{\downarrow\uparrow})$$
$$+\rho_0(E)\frac{\tau^+}{2}P_\tau(W_{\uparrow\uparrow}-W_{\downarrow\downarrow}+W_{\uparrow\downarrow}-W_{\downarrow\uparrow}), \tag{B2}$$

and

$$F_{\uparrow\uparrow}+F_{\downarrow\downarrow}-F_{\downarrow\uparrow}-F_{\uparrow\downarrow}$$
$$=\rho_0(E)\frac{\tau^+}{2}(W_{\uparrow\uparrow}+W_{\downarrow\downarrow}-W_{\downarrow\uparrow}-W_{\uparrow\downarrow})$$
$$+\rho_0(E)\frac{\tau^+}{2}P_\tau(W_{\uparrow\uparrow}-W_{\downarrow\downarrow}+W_{\downarrow\uparrow}-W_{\uparrow\downarrow})\ , \tag{B3}$$

with the transition probabilities $W_{\sigma'\sigma}$ defined in Eq. (14).

In light of our lowest-order approximation we only need the first terms in Eqs. (B2) and (B3), but both of them in Eq. (B1). In addition we replace $\psi^+=(N_\uparrow/\tau_\uparrow)+(N_\downarrow/\tau_\downarrow)$ by its lowest-order paramagnetic result ($\uparrow=\downarrow$) N/τ and τ^+ by 2τ (note N does not contain source function electrons). This yields for $\Delta(E)$

$$\Delta(E)=\Delta_s(E)+\frac{\int dE'\,N(E')(W_{\uparrow\uparrow}-W_{\uparrow\uparrow})\Delta(E')}{\int dE'\,N(E')(W_{\uparrow\uparrow}+W_{\uparrow\downarrow})}$$
$$+R_1+R_2\ , \tag{B4}$$

where $\Delta_s(E)$ is dealt with in Appendix D and where the remainder terms R_1 and R_2 are defined as

$$R_1=\frac{F(E_0,E)-p_BD(E_0,E)}{2\bar\psi^+(E)} \tag{B5}$$

and

$$R_2=\int dE'\,N(E')[W_{\uparrow\uparrow}-W_{\downarrow\downarrow}+W_{\downarrow\uparrow}-W_{\uparrow\downarrow}$$
$$-2p_B(W_{\downarrow\uparrow}+W_{\uparrow\downarrow})]\rho_0(E)/2\bar\psi^+(E)\ . \tag{B6}$$

R_1 and R_2 will be shown to be negligible provided there is electron multiplication and that the matrix elements involved have a certain relationship. We start with R_1. Extracting $D(E_0,E)$, we can write R_1 as

$$R_1=\frac{D(E_0,E)}{2\bar\psi^+(E)}[p_s(E_0,E)-p_B]\ , \tag{B7}$$

where

$$p_s(E',E)=F(E',E)/D(E',E)$$

is the polarization at E created in a single scattering event by an electron initially at E' provided E' is sufficiently large that the lifetime difference between $\tau_\uparrow$ and $\tau_\downarrow$ can be neglected. Now because of the electron multiplication we have from Eq. (41) that $2\bar\psi^+(E)\gg D(E_0,E)$ which means that $R_1\ll[p_s(E_0,E)-p_B]$. Now $p_s(E_0,E)$ the single scattering probability is constructed in such a way that $|p_s(E_0,E)|\le 1$ which means that $p_s(E_0,E)-p_B$ is at most of $O(1)$.

In the case of the simple model studied in Sec. V, $R_1=R_1^0$ is given by

$$R_1^0=-\tfrac{1}{2}p_B(E/E_0)^2\ , \tag{B8}$$

which indeed is much less than p_B for $E\ll E_0$. In Appendix C the term R_2 is also shown to be negligible compared to p_B yielding Eq. (43a) for $\Delta(E)$.

APPENDIX C

We want to show that R_2 is small compared to p_B:

$$R_2=\int_E^{E_0} dE' N(E')[W_{\uparrow\uparrow}-W_{\downarrow\downarrow}+W_{\downarrow\uparrow}-W_{\uparrow\downarrow} -2p_B(W_{\downarrow\uparrow}+W_{\uparrow\downarrow})]\rho_0(E)/2\bar{\psi}^+ . \tag{C1}$$

In order to place things on an equal footing we replace $\bar{\psi}^+$ with $\frac{1}{2}\int dE'\bar{\psi}^+(E')D(E',E)$ from the multiple scattering [see Appendix B and Eq. (41)]. Rewritten in terms of W's and working in the paramagnetic limit, we have

$$\bar{\psi}^+(E)\simeq\int dE' N(E')(W_{\uparrow\uparrow}+W_{\uparrow\downarrow})\rho_0(E) . \tag{C2}$$

This term is also used for $\bar{\psi}^+(E)$ to arrive at Eq. (43a) for Δ.

With the expressions for $W(E'\sigma,E\sigma)$ in Eq. (14) we can write

$$W_{\uparrow\uparrow}-W_{\downarrow\downarrow}+W_{\downarrow\uparrow}-W_{\uparrow\downarrow}=\frac{2\pi}{\hbar}(A_1+A_2) , \tag{C3}$$

where

$$A_1=2\int d\varepsilon[\rho_\uparrow^<(\varepsilon)-\rho_\downarrow^<(\varepsilon)]\rho_0^>(\varepsilon+\omega)\,|M_{ss}^{sd}(E-\varepsilon)|^2 \tag{C4}$$

and

$$A_2=\int d\varepsilon[\rho_\uparrow^<(\varepsilon)-\rho_\downarrow^<(\varepsilon)]\rho_d^>(\varepsilon+\omega)\,|M_{ds}^{sd}(E'-E_F)|^2 . \tag{C5}$$

We have used the fact that $\rho_\uparrow^>=\rho_\downarrow^>\equiv\rho_0(\varepsilon)$ when ε is above the spin-down d-band peak and that $\rho_\downarrow^>=\rho_d^>(\varepsilon)+\rho_0^>(\varepsilon)$ when ε is below, while $\rho_\uparrow^<$ stays unchanged. Also $M_{ds}^{sd}(E'-\varepsilon)\simeq M_{ds}^{sd}(E'-E_F)$ since $\rho_d\neq 0$ only very close to E_F.

In the same fashion we obtain

$$W_{\downarrow\uparrow}+W_{\uparrow\downarrow}=\frac{2\pi}{\hbar}\int d\varepsilon[\rho_\uparrow^<(\varepsilon)+\rho_\downarrow^<(\varepsilon)]\rho_0^>(\varepsilon+\omega) \times|M_{ss}^{sd}(E-\varepsilon)|^2+\frac{2\pi}{\hbar}A_3 ,$$

where

$$A_3\equiv\int d\varepsilon\rho_\uparrow^<(\varepsilon)\rho_d^>(\varepsilon+\omega)\,|M_{ds}^{sd}(E'-E_F)|^2 . \tag{C6}$$

Dividing R into the regular (ρ_0) and the "irregular" (ρ_d) piece writing $R=R_0+R_d$ we have for R_0

$$R_0=\left[\frac{2\pi}{\hbar}\int dE' N(E')\left[\int d\varepsilon[\rho_\uparrow^<(\varepsilon)-\rho_\downarrow^<(\varepsilon)]\rho_0^>(\varepsilon+\omega)\,|M_{ss}^{sd}(E-\varepsilon)|^2 - p_B\int d\varepsilon[\rho_\uparrow^<(\varepsilon)+\rho_\downarrow^<(\varepsilon)]\rho_0^>(\varepsilon+\omega)\,|M_{ss}^{sd}(E-\varepsilon)|^2\right]\right]\Big/\left[\int dE' N(E')(W_{\uparrow\uparrow}+W_{\uparrow\downarrow})\right] . \tag{C7}$$

For the simple model ρ_0 and M_{ss}^{sd} are constants and we see that R_0 vanishes identically. But even for slowly varying functions R_0 has to be very small. $\rho_\uparrow^<\pm\rho_\downarrow^<$ only exists over the bandwidth W and with no significant change in ρ_0 ($\sim\sqrt{\varepsilon}$) and $M_{ss}^{sd}(E-\varepsilon)$ over an energy scale of W we can safely neglect R_0, Eq. (C7). Or to turn it the other way. In case the final free-electron density of states or the matrix element M_{ss}^{sd} has a major variation over the bandwidth of the solid we do not expect, in general, that R_0 has to be small and in this case ψ^-/ψ^+ cannot be expected to stay close to p_B.

Since ρ_d is a very narrow function in energy we can approximate A_2 and A_3 as

$$A_2\simeq[\rho_\uparrow^<(-\omega)-\rho_\downarrow^<(-\omega)]\,|M_{ds}^{sd}(E'-E_F)|^2\int d\varepsilon\rho_d^>(\varepsilon+\omega) \tag{C8}$$

and

$$A_3\simeq\rho_\uparrow^<(-\omega)\,|M_{ds}^{sd}(E'-E_F)|^2\int d\varepsilon\rho_d^>(\varepsilon+\omega) , \tag{C9}$$

where we notice that for $\omega>W$ they both vanish. Now $\int d\varepsilon\rho_d^>(\varepsilon+\omega)\equiv n_\uparrow-n_\downarrow$ by definition. Changing the integration in Eq. (C1) to go over every transfer ω we can write R_d as

$$R_d=\frac{\int_{-W}^0 d\omega N(E-\omega)\,|M_{ds}^{sd}(E-E_F-\omega)|^2[\rho_\uparrow^<(\omega)-\rho_\downarrow^<(\omega)-2p_B\rho_\uparrow^<(\omega)](n_\uparrow-n_\downarrow)}{2\int d\varepsilon\int_E^{E_0} dE' N(E')[\rho_\uparrow^<(\varepsilon)+\rho_\downarrow^<(\varepsilon)][\,|M_{ss}^{sd}(E'-E)|^2+|M_{ss}^{sd}(E-\varepsilon)|^2]\rho_0^>(\varepsilon+\omega)} , \tag{C10}$$

neglecting the ρ_d contribution in the denominator, working in the paramagnetic limit. We immediately see from its structure that it scales with M_{ds}^{sd}/M_{ss}^{sd} (they are both evaluated at a very low energy), so if this ratio is small R_d is indeed small compared to p_B.

Because $N(E)$ strongly weights small E we can take out all the matrix elements evaluated at a very small energy, provided they do not vary strongly over W (the ε integration). Performing the integration over the bandwidth and extracting the least favorable $N(E)$ gives

$$|R_d|<\frac{n_\uparrow-n_\downarrow}{4W\rho_0(E_F)}\,\frac{N(E)}{N(E+W)}\left|\frac{M_{ds}^{sd}}{M_{ss}^{sd}}\right|^2 p_B^2\sim\left|\frac{M_{ds}^{sd}}{M_{ss}^{sd}}\right|^2 p_B , \tag{C11}$$

where the estimate is for $N\sim E^{-3}$ with $E\sim W$, $2W\rho_0(E_F)\sim 1$ and $p_B\sim 10\%$. Thus as long as $M_{ds}^{sd}(0)$ is negligible compared with $M_{ss}^{sd}(0)R_d$ is small compared to p_B. Since $R_d\sim O(p_B^3)$ it should work better for Ni than for Fe.

APPENDIX D

We will justify the form of the source function in Eq. (43b). From Appendix B we have

$$\Delta_s(E)=\frac{\int dE' N(E')P_\tau(E')(W_{\uparrow\uparrow}-W_{\uparrow\downarrow})\rho_0(E)}{\bar{\psi}^+(E)} . \quad \text{(D1)}$$

Using the electron multiplication to replace $\bar{\psi}^+$ by $\frac{1}{2}\int dE'\bar{\psi}^+(E')D(E',E)$ we can write

$$\Delta_s(E)=\frac{\int dE' N(E')P_\tau(E')(W_{\uparrow\uparrow}+W_{\uparrow\downarrow})}{2\int dE' N(E')(W_{\uparrow\uparrow}+W_{\uparrow\downarrow})}+R_3 , \quad \text{(D2)}$$

where

$$R_3=\frac{\int dE' N(E')P_\tau(E')(W_{\uparrow\uparrow}-3W_{\uparrow\downarrow})}{2\int dE' N(E')(W_{\uparrow\uparrow}+W_{\uparrow\downarrow})} . \quad \text{(D3)}$$

Now $W_{\uparrow\uparrow}-3W_{\uparrow\downarrow}$ is equal to [cf. Eq. (14)]

$$\begin{aligned} W_{\uparrow\uparrow}-3W_{\uparrow\downarrow}= &\int d\varepsilon\rho_\uparrow^<(\varepsilon)\rho_0^>(\varepsilon+\omega) \\ &\times[\,|M(\omega)|^2+|M(E-\varepsilon)|^2] \\ &+\int d\varepsilon\rho_\downarrow^<(\varepsilon)\rho_0^>(\varepsilon+\omega) \\ &\times[\,|M(\omega)|^2-3|M(E-\varepsilon)|^2] , \end{aligned} \quad \text{(D4)}$$

where we stay in the paramagnetic limit because P_τ is already a lowest-order quantity. We can rewrite Eq. (D3) as

$$\begin{aligned} W_{\uparrow\uparrow}-3W_{\uparrow\downarrow}= &\int d\varepsilon[\rho_\uparrow^<(\varepsilon)-\rho_\downarrow^<(\varepsilon)]\rho_0^>(\varepsilon+\omega) \\ &\times[\,|M(\omega)|^2+|M(E-\varepsilon)|^2] \\ &+2\int d\varepsilon\rho_\downarrow^<(\varepsilon)\rho_0^>(\varepsilon+\omega) \\ &\times[\,|M(\omega)|^2-|M(E-\varepsilon)|^2] . \end{aligned} \quad \text{(D5)}$$

Again for a model where $\rho_0 M$ is constant or varies slowly over the bandwidth this gives $W_{\uparrow\uparrow}-3W_{\uparrow\downarrow}\propto p_B$ and R_3 is of second order in p_B. Because of the strong weighting of N and P_τ toward lower energies both $\omega=E'-E$ and $E-\varepsilon$ will be small energies and $M(\omega)$ must be very close to $M(E-\varepsilon)$. In the same spirit the first term in Eq. (D4) is of first order in $\uparrow$ minus $\downarrow$ and multiplied by P_τ becomes of second order in small quantities, the other factor being M_{ds}^{sd}/M_{ss}^{sd} and should consequently be dropped.

APPENDIX E

In this appendix we will write down the expression for P_τ in terms of the basic densities of states and matrix elements we have been working with. P_τ is defined as

$$P_\tau=\frac{\tau_\uparrow-\tau_\downarrow}{\tau_\uparrow+\tau_\downarrow}\equiv\frac{(1/\tau_\downarrow)-(1/\tau_\uparrow)}{(1/\tau_\downarrow)+(1/\tau_\uparrow)} . \quad \text{(E1)}$$

Using Eq. (24) we find

$$\begin{aligned} \frac{1}{\tau_\downarrow}-\frac{1}{\tau_\uparrow}=\frac{2\pi}{\hbar}\int_{E_F}^{E'}dE\,\rho_d^>(E)\int d\varepsilon[\rho_\uparrow^<(\varepsilon)+\rho_\downarrow^<(\varepsilon)] \\ \times\rho_0^>(\varepsilon+\omega)\,|M_{ds}^{sd}(\omega)|^2 . \end{aligned} \quad \text{(E2)}$$

For the ferromagnetic materials ρ_d is a very sharply peaked function just above the Fermi level yielding

$$\begin{aligned} \frac{1}{\tau_\downarrow}-\frac{1}{\tau_\uparrow}\simeq\frac{2\pi}{\hbar}(n_\uparrow-n_\downarrow)\,|M_{ds}^{sd}(E'-E_F)|^2 \\ \times[\bar{\rho}_{\uparrow,s}(E'-E_F)+\bar{\rho}_{\downarrow,s}(E'-E_F)] , \end{aligned} \quad \text{(E3)}$$

$\bar{\rho}_{\sigma,s}$ being the joint density of states [Eq. (29)], and we have used $\int d\varepsilon\rho_d(\varepsilon)\equiv n_\uparrow-n_\downarrow$, when E' is outside of the peak. $(1/\tau_\uparrow)+(1/\tau_\downarrow)$ can be evaluated in the paramagnetic limit and with the same approximations as above we get

$$\begin{aligned} \frac{1}{\tau_\downarrow}+\frac{1}{\tau_\uparrow}\simeq\frac{2\pi}{\hbar}2\int_{E_F}^{E'}dE\,\rho_0^>(E)\int d\varepsilon[\rho_\uparrow^<(\varepsilon)+\rho_\downarrow^<(\varepsilon)] \\ \times\rho_0^>(\varepsilon+\omega)\,|M_{ss}^{sd}(\omega)|^2 \\ =\frac{4\pi}{\hbar}\int_{E_F}^{E'}dE\,\rho_0^>(E)\,|M_{ss}^{sd}(E'-E)|^2 \\ \times[\bar{\rho}_{\uparrow,s}(E'-E)+\bar{\rho}_{\downarrow,s}(E'-E)] . \end{aligned} \quad \text{(E4)}$$

and clearly P_τ is strongly related to M_{ds}^{sd}/M_{ss}^{sd}.

[1]J. Unguris, D. T. Pierce, A. Galejs, and R. J. Celotta, Phys. Rev. Lett. **49**, 72 (1982).
[2]E. Kisker, W. Gudat, and K. Schröder, Solid State Commun. **44**, 591 (1982).
[3]H. Hopster, R. Raue, E. Kisker, G. Güntherodt, and M. Campagna, Phys. Rev. Lett. **50**, 70 (1983).
[4]M. Landolt and D. Mauri, Phys. Rev. Lett. **59**, 1783 (1982).
[5]J. Glazer, Ph.D. thesis, University of Trieste, 1984.
[6]D. R. Penn, S. P. Apell, and S. M. Girvin, Phys. Rev. Lett. **55**, 518 (1985).
[7]J. L. Erskine and E. A. Stern, Phys. Rev. Lett. **30**, 1329 (1973).
[8]P. A. Wolff, Phys. Rev. **95**, 56 (1954).
[9]R. E. Marshak, Rev. Mod. Phys. **19**, 185 (1947).
[10]C. N. Berglund and W. E. Spicer, Phys. Rev. **136**, A1030 (1964).
[11]E. O. Kane, Phys. Rev. **159**, 624 (1967).
[12]R. W. Rendell and David R. Penn, Phys. Rev. Lett. **45**, 2057 (1980).
[13]V. L. Moruzzi, J. F. Janak, and A. R. Williams, *Calculated Electronic Properties of Metals* (Pergamon, New York, 1978).
[14]J. Glazer, E. Tosatti, H. Hopster, R. Kurzawa, W. Schmitt, K. H. Walker, and G. Güntherodt (unpublished).
[15]J. A. D. Matthew, Phys. Rev. B **25**, 3326 (1982).
[16]A. Bringer, M. Campagna, R. Feder, W. Gudat, E. Kisker,

and E. Kulhmann, Phys. Rev. Lett. **42**, 1705 (1979).
[17]H. Hopster, R. Raue, and R. Clauberg, Phys. Rev. Lett. **53**, 695 (1984).
[18]J. Kirschner, D. Rebenstorff, and H. Ibach, Phys. Rev. Lett. **53**, 698 (1984).
[19]E. Kisker, W. Gudat, M. Campagna, E. Kuhlmann, H. Hopster, and I. D. Moore, Phys. Rev. Lett. **43**, 966 (1979).
[20]R. H. Victora, L. M. Falicov, and S. Ishida, Phys. Rev. **30**, 3896 (1984).
[21]J. Glazer and E. Tosatti, Solid State Commun. **52**, 905 (1984).