Low-energy electron mean free path and its spin dependence in transition metals

Henri-Jean Drouhin
Laboratoire de Physique de la Matière Condensée, Ecole Polytechnique, 91128 Palaiseau, France
(Received 4 September 1997)

A simple model is presented to estimate the low-energy electron mean free path in transition metals and to calculate its spin dependence. In this model, the electron-scattering rate is directly related to the number of holes in the *d* bands; its spin asymmetry is almost proportional to the spin magnetic moment. These quantities show a weak energy dependence from the vacuum level up to a few tens of eV above the metal Fermi level. [S0163-1829(97)04947-3]

The study of the inelastic electron mean free path (IMFP) in metals started many years ago, both theoretically[1] and experimentally.[2] The high-energy domain (say for electron energies from 50 eV up to a few keV above the Fermi level) now seems well understood; the question of the low-energy electron mean free path was considered to remain puzzling, because in this energy domain the electrons are sensitive to the details of the band structure, and because, there, several energy-loss mechanisms can be involved. It became clear that, at very low energy, close to zero, the electron mean free path is extremely large (tens of nm from the conductivity relaxation time), whereas it reaches a minimum value (a few tenths of a nm) at energies of the order of 100 eV. Its dependence on the electron energy was thought to follow a ''universal'' curve.[3] However, near the vacuum level (typically 4 eV), the scarce experimental data all originated from the work by Kanter.[4] Recent experiments indicated that the IMFP was probably more material dependent than previously assumed,[5] and in particular the large increase of the IMFP expected when the electron energy is lowered close to the vacuum level does not seem to be observed in transition metals.[6] At the same time, the spin dependence of the IMFP was considered,[7] and, for example in the case of cobalt, a very high spin asymmetry was measured at a few eV above the Fermi level,[8] the asymmetry remaining significant at higher electron energy.[9] An important step was the proposition by Schönhense and Siegmann of an empirical model in which the scattering cross section, proportional to the inverse of the IMFP, is directly related to the number of holes in the *d* bands.[5] A consequence is that the quantity $(1/\lambda^-)-(1/\lambda^+)$, where λ^- (λ^+) is the IMFP for minority (majority) electrons, is proportional to the spin magnetic moment. The spin dependence of the electron mean free path at the Fermi level is also crucial in the analysis of giant magnetoresistance experiments. In this paper, we study the low-energy IMFP (in fact, the scattering rate) and its spin dependence for electron energies ranging from zero up to a few tens of eV, with elementary assumptions but using a realistic description of the d-band structure.

We only take into account electron-electron scattering, and consider a primary electron at energy E_p (hereafter the energy origin is set at the metal Fermi level), with a spin $\sigma=\pm$. This electron loses an amount of energy ε, which is used to excite a secondary electron from a negative energy to a positive energy ε', with spin conservation. The transition rate $r^\sigma(\varepsilon)$ from E_p to $(E_p-\varepsilon)$ for the primary electron is taken as (random-**k** approximation)

$$r^\sigma(\varepsilon)=n^\sigma(E_p-\varepsilon)[\omega^{\sigma\sigma}\phi^{\sigma\sigma}(\varepsilon)+\omega^{\sigma-\sigma}\phi^{-\sigma-\sigma}(\varepsilon)],$$

$$\phi^{\sigma\sigma'}(\varepsilon)=\int_0^\varepsilon n^\sigma(\varepsilon')n^{\sigma'}(\varepsilon'-\varepsilon)d\varepsilon'.$$

In the expression of $r^\sigma(\varepsilon)$, $\omega^{\sigma\sigma}$ and $\omega^{\sigma-\sigma}$ are taken as constant transition matrix elements characterizing direct and exchange processes; for simplicity, we also assume that we have the symmetry relations $\omega^{++}=\omega^{--}=\omega$ and $\omega^{+-}=\omega^{-+}=\omega_f$. The functions $n^\sigma(u)$ are the relevant densities of states. The $\phi^{\sigma\sigma'}(\varepsilon)$ functions are defined for $\sigma'=\pm\sigma$, an extension which will be useful later. Two important parameters can be introduced: the spin-averaged transition rate $\mathcal{R}(E_p)=\frac{1}{2}[R^-(E_p)+R^+(E_p)]$, and the spin-dependent component of the transition rates, $\Delta\mathcal{R}(E_p)=\frac{1}{2}[R^-(E_p)-R^+(E_p)]$; the spin-dependent scattering rate at energy E_p, $R^\sigma(E_p)$ is

$$R^\sigma(E_p)=\int_0^{E_p}r^\sigma(\varepsilon)d\varepsilon.$$

Note that the ratio $\Delta\mathcal{R}(E_p)/\mathcal{R}(E_p)=(\lambda^+-\lambda^-)/(\lambda^++\lambda^-)$ is the standard spin asymmetry of the mean free path.

Hereafter, the band structure is described by a constant density of states n_{sp}, corresponding to the density of states at the Fermi level for s and p bands on which is superimposed a positive continuous compact-support function $n_d^\sigma(u)$ describing the considered d band: $n^\sigma(u)=n_d^\sigma(u)+n_{sp}$. The d bands are bounded by energies E_1^σ (lower bound) and E_2^σ (upper bound); their common width is denoted as W_d. The numbers N_e^σ and N_h^σ of electrons and holes in the d bands ($N_e^\sigma+N_h^\sigma=5$) are

$$N_e^\sigma=\int_{-\infty}^0 n_d^\sigma(u)du,\quad N_h^\sigma=\int_0^{+\infty}n_d^\sigma(u)du.$$

The band centers E_d^σ and the centers of the emerged part of the d bands, E_h^σ, are defined as

$$E_d^\sigma=\frac{\int_{-\infty}^{+\infty}un_d^\sigma(u)du}{\int_{-\infty}^{+\infty}n_d^\sigma(u)du}=\frac{1}{5}\int_{-\infty}^{+\infty}un_d^\sigma(u)du,$$

$$E_h^\sigma N_h^\sigma = \int_0^{+\infty} u n_d^\sigma(u) du.$$

It can be easily shown that the $\phi^{\sigma\sigma'}(\varepsilon)$ functions verify the following properties:

(i) For small ε, $\phi^{\sigma\sigma'}(\varepsilon) \sim n^\sigma(0) n^{\sigma'}(0)\varepsilon$.

(ii) For large values of ε $[\varepsilon \geqslant W^{\sigma\sigma'} = \mathrm{Sup}(|E_1^\sigma|,|E_2^\sigma|,|E_1^{\sigma'}|,|E_2^{\sigma'}|,|E_2^\sigma - E_1^{\sigma'}|,|E_2^{\sigma'} - E_1^\sigma|)]$, we find

$$\phi^{\sigma\sigma'}(\varepsilon) = n_{sp}^2 \varepsilon + n_{sp}(N_h^\sigma + N_e^{\sigma'}).$$

A consequence is that the value of $\phi^{\sigma\sigma}(\varepsilon)$ does not depend on the band considered; also note that, if $n_{sp}=0$, $\phi^{\sigma\sigma'}(\varepsilon) = 0$ when $\varepsilon \geqslant \mathrm{Sup}(|E_2^\sigma - E_1^{\sigma'}|,|E_2^{\sigma'} - E_1^\sigma|)$.

(iii) When $u \geqslant \mathrm{Sup}(|E_2^\sigma - E_1^{\sigma'}|,|E_2^{\sigma'} - E_1^\sigma|)$,

$$\int_0^u \phi_d^{\sigma\sigma'}(\varepsilon) d\varepsilon = N_h^\sigma N_e^{\sigma'},$$

where $\phi_d^{\sigma\sigma'}(\varepsilon)$ is $\phi^{\sigma\sigma'}(\varepsilon)$ calculated for the only d bands, i.e., with $n_{sp}=0$.

For very small primary energies, $R^\sigma(E_p)$ is easily evaluated [using (i)], and we obtain

$$\tfrac{1}{2}[R^-(E_p) \pm R^+(E_p)] = \tfrac{1}{4} E_p^2 [n^-(0) \pm n^+(0)] \times \{\omega[n^-(0)^2 + n^+(0)^2] \pm (\omega_f - \omega) n^-(0) n^+(0)\}.$$

If $\omega = \omega_f$, or if $n^+(0)=0$ (perfect ferromagnet), or if $n^+(0)=n^-(0)$ (nonmagnetic metal), we conclude that the spin asymmetry of the IMFP is the asymmetry in the density of states at the Fermi level:

$$\frac{\Delta\mathcal{R}(E_p)}{\mathcal{R}(E_p)} = \frac{n^-(0) - n^+(0)}{n^-(0) + n^+(0)}.$$

For a large primary energy $[E_p \geqslant \mathrm{Sup}(W^{++},W^{--}) + \mathrm{Sup}(0,\ E_2^-,E_2^+)]$, $R^\sigma(E_p)$ is calculated after cutting the integration domain at energy $W = \mathrm{Sup}(W^{++},W^{--})$:

$$R^\sigma(E_p) = \int_0^{E_p} r^\sigma(\varepsilon) d\varepsilon = \int_0^W r^\sigma(\varepsilon) d\varepsilon + \int_W^{E_p} r^\sigma(\varepsilon) d\varepsilon.$$

When $0 \leqslant \varepsilon \leqslant W$, $(E_p - \varepsilon)$ lies out of the d bands, so that $n^\sigma(E_p - \varepsilon) = n_{sp}$. Considering only $\phi_d^{\sigma\sigma}(\varepsilon)$ in the expression of $\phi^{\sigma\sigma}(\varepsilon)$, the first integral yields [using (iii)]

$$n_{sp}(\omega N_e^\sigma N_h^\sigma + \omega_f N_e^{-\sigma} N_h^{-\sigma}).$$

The other terms are easily calculated after an integration by parts. Concerning the domain $W \leqslant \varepsilon \leqslant E_p$, we use the large-energy expression of $\phi^{\sigma\sigma}(\varepsilon)$ [see (ii)], so that the corresponding integral reads

$$n_{sp}(\omega + \omega_f) \int_W^{E_p} n^\sigma(E_p - \varepsilon)[n_{sp}\varepsilon + 5] d\varepsilon.$$

Finally, the expression of $R^\sigma(E_p)$ reduces to

$$R^\sigma(E_p) = n_{sp}(\omega N_e^\sigma N_h^\sigma + \omega_f N_e^{-\sigma} N_h^{-\sigma}) + 5 n_{sp}^2(\omega E_d^\sigma + \omega_f E_d^{-\sigma}) - 2 n_{sp}^2(\omega E_h^\sigma N_h^\sigma + \omega_f E_h^{-\sigma} N_h^{-\sigma}) + n_{sp}(\omega + \omega_f)\left[(5 - n_{sp}E_h^\sigma) N_h^\sigma + 5 n_{sp} E_p \times \left(1 + \frac{N_h^\sigma}{5}\right) + \tfrac{1}{2} n_{sp}^2 E_p^2\right].$$

To express $\mathcal{R}(E_p)$ and $\Delta\mathcal{R}(E_p)$, we introduce the total hole number $\mathcal{N}_h = (N_h^- + N_h^+)$ and $\Delta\mathcal{N}_h = (N_h^- - N_h^+)$. Moreover, we define $\gamma\mathcal{N}_h = [(N_h^-)^2 + (N_h^+)^2]$ and the characteristic energies $E_d = -(E_d^- + E_d^+)/2$, $E_h \mathcal{N}_h = (N_h^- E_h^- + N_h^+ E_h^+)$, $E_h' \Delta\mathcal{N}_h = (N_h^- E_h^- - N_h^+ E_h^+)$, and $E_{\mathrm{ex}} = (E_d^- - E_d^+)$; then

$$\mathcal{R}(E_p) = 5 n_{sp}(\omega + \omega_f)\left[\mathcal{N}_h\left(1 - \frac{\gamma}{10} + \frac{1}{10} n_{sp}(E_p - 3E_h)\right) + n_{sp}(E_p - E_d) + \frac{1}{10} n_{sp}^2 E_p^2\right]$$

and

$$\Delta\mathcal{R}(E_p) = \tfrac{5}{2} n_{sp}(\omega - \omega_f)[\Delta\mathcal{N}_h(1 - \tfrac{1}{5}\mathcal{N}_h - \tfrac{2}{5} n_{sp} E_h') + n_{sp} E_{\mathrm{ex}}] + \tfrac{5}{2} n_{sp}(\omega + \omega_f)\Delta\mathcal{N}_h[1 + \tfrac{1}{5} n_{sp}(E_p - E_h')].$$

An important conclusion is that $\mathcal{R}(E_p)$ is determined by the number of holes in the d bands, as postulated in the Schönhense and Siegmann model.[5] When $\mathcal{N}_h \neq 0$, the relation $\mathcal{N}_h = n_{sp} E^*$ defines an energy scale which is relevant for a significant variation of $\mathcal{R}(E_p)$. Because n_{sp} is almost ten times smaller than the mean density of states in the d bands $(5/W_d)$, $E^* \sim 2 W_d \mathcal{N}_h$. Assuming that none of the d bands is empty, and using a square-band model, we find $E_h = \gamma(W_d/10)$, $E_h' = \mathcal{N}_h(W_d/10)$, and $E_{\mathrm{ex}} \sim \Delta\mathcal{N}_h(W_d/5)$. As expected, $\Delta\mathcal{R}(E_p)$ appears to be proportional to $\Delta\mathcal{N}_h$, i.e., to the spin magnetic moment. For a material with a small hole number, if we retain only the leading terms in the expression of $\mathcal{R}(E_p)$ and $\Delta\mathcal{R}(E_p)$, we obtain

$$\mathcal{R}(E_p) \approx 5 n_{sp}(\omega + \omega_f)\mathcal{N}_h\left[1 + \frac{E_p - E_d}{E^*} + \frac{\mathcal{N}_h}{10}\left(\frac{E_p}{E^*}\right)^2\right],$$

$$\Delta\mathcal{R}(E_p) \approx 5 n_{sp}\omega\Delta\mathcal{N}_h\left[1 + \frac{\mathcal{N}_h}{10}\left(1 + \frac{\omega_f}{\omega}\right)\left(\frac{\omega_f - \omega}{\omega_f + \omega} + \frac{E_p}{E^*}\right)\right].$$

To simplify the discussion, we now assume that $\omega \sim \omega_f$ (see below). Then we observe that most of the spin dependence of the IMFP involves large energy losses, the primary electron falling in the d bands. Concerning the scattering rate, a comparable contribution originates from small energy losses, the secondary electron being excited inside the d states. In the case of Fe, Co, or Ni, because $N_h^+ \ll N_h^-$, $\Delta\mathcal{N}_h/\mathcal{N}_h \approx 1$. In all of these cases, we estimate $\Delta\mathcal{R}/\mathcal{R} \sim 0.4$ when $E_p \sim W_d \sim 6$ eV, i.e., close to the vacuum level, although the bulk polarizations $p_B = \Delta\mathcal{N}_h/(10 - \mathcal{N}_h)$ are strongly different

[27% for Fe, 17% for Co, and 6% for Ni].[7] The energy dependence of the spin asymmetry is almost governed by the energy dependence of $\mathcal{R}(E_p)$. Note that, to convert a scattering time into an IMFP, we have to multiply it by the electron velocity. If the sp band is described in a parabolic model, the electron velocity is proportional to $[E_p+E_F]^{1/2}$, where $E_F\sim 10$ eV is the Fermi energy; this is is not strictly consistent with a constant n_{sp}, which implies an electron velocity proportional to $[E_p+E_F]^{2/3}$. In any case, the IMFP does not vary much with energy.

As previously stated, the spin polarization of the secondary electrons involves the matrix elements ω and ω_f. The excitation at energy ε' of secondary electrons with a spin σ' by primary electrons of spin σ occurs at the rates

$$\rho^{\sigma\sigma'}(\varepsilon')=\omega^{\sigma\sigma'}n^{\sigma'}(\varepsilon')\int_{\varepsilon'}^{E_p}n^{\sigma}(E_p-\varepsilon)n^{\sigma'}(\varepsilon'-\varepsilon)d\varepsilon$$

$$=\omega^{\sigma\sigma'}n^{\sigma'}(\varepsilon')\int_0^{E_p-\varepsilon'}$$

$$\times n^{\sigma}(u)n^{\sigma'}[u-(E_p-\varepsilon')]du,$$

$$\rho^{\sigma\sigma'}(\varepsilon')=\omega^{\sigma\sigma'}n^{\sigma'}(\varepsilon')\phi^{\sigma\sigma'}(E_p-\varepsilon').$$

We consider the energy domain Sup(0, $E_2^{\sigma},E_2^{\sigma'})\leqslant\varepsilon'\leqslant E_p-W^{-+}$. There, $n^-(\varepsilon')=n^+(\varepsilon')=n_{sp}$, and we start with a primary beam with a spin polarization P; the numbers of primary electrons with spin + or − are $(1+P)/2$ and $(1-P)/2$, respectively. The spin polarization $\mathcal{P}$ of the secondary electrons is then:

$$\mathcal{P}=\frac{\omega(\phi^{++}-\phi^{--})+\omega_f(\phi^{-+}-\phi^{+-})+P[\omega(\phi^{++}+\phi^{--})-\omega_f(\phi^{+-}+\phi^{-+})]}{\omega(\phi^{++}+\phi^{--})+\omega_f(\phi^{+-}+\phi^{-+})+P[\omega(\phi^{++}-\phi^{--})+\omega_f(\phi^{+-}-\phi^{-+})]},$$

with [using (ii)]

$$\phi^{++}(E_p-\varepsilon')-\phi^{--}(E_p-\varepsilon')=0,$$

$$\phi^{++}(E_p-\varepsilon')+\phi^{--}(E_p-\varepsilon')=\phi^{+-}(E_p-\varepsilon')+\phi^{-+}(E_p-\varepsilon')=10n_{sp}[1+\tfrac{1}{5}n_{sp}(E_p-\varepsilon')],$$

$$\phi^{-+}(E_p-\varepsilon')-\phi^{+-}(E_p-\varepsilon')=2n_{sp}\Delta\mathcal{N}_h,$$

so that

$$\mathcal{P}\approx\frac{\omega_f p_B\left(1-\frac{\mathcal{N}_h}{10}\right)+P\,\frac{(\omega-\omega_f)}{2}\,[1+\frac{1}{5}n_{sp}(E_p-\varepsilon')]}{\frac{(\omega+\omega_f)}{2}\,[1+\frac{1}{5}n_{sp}(E_p-\varepsilon')]-\omega_f P p_B\left(1-\frac{\mathcal{N}_h}{10}\right)}.$$

Unless $\omega\sim\omega_f$, the spin polarization of the secondary electrons has a strong memory of the primary beam polarization. Note that the primary electrons of spin σ emerge at energy ε', after suffering a collision, at the rate

$$r^{\sigma}(E_p-\varepsilon')=n^{\sigma}(\varepsilon')[\omega\phi^{\sigma\sigma}(E_p-\varepsilon')$$
$$+\omega_f\phi^{-\sigma-\sigma}(E_p-\varepsilon')].$$

For a nonmagnetic metal, the distribution of the primary electrons which have lost energy always coincides with the distribution of the secondary electrons. For a ferromagnetic metal, in the energy domain considered above, the total number of primary electrons at energy ε' is $(1/2)n_{sp}(\omega+\omega_f)[\phi^{++}(E_p-\varepsilon')+\phi^{--}(E_p-\varepsilon')]$, which is almost equal to the number of secondary electrons when $P\Delta\mathcal{N}_h/5\ll 1$.

The present model provides a simple derivation and generalization of the empirical Schönhense and Siegmann relation between the scattering cross section and the number of holes in the d bands.[5] The large spin asymmetries (between 0.6 and 0.4) reported for Co in Ref. 8 at a few eV, the value measured at 14 eV ($\sim$0.15),[9] the comparable asymmetries reported near the vacuum level for Fe[5,10] as well as the weak IMFP and asymmetry variations versus energy also reported for Fe in Ref. 6 (asymmetry divided by a factor of the order of 2 between 10- and 40-eV primary energy) agree nicely with this model. It allows us to understand, at least qualitatively, why a negligible spin asymmetry in the mean free path was reported for Fe in spin-valve experiments,[11] which involve phenomena arising at the Fermi level: a cancellation occurs due to the presence of empty d states in the majority band. A definite conclusion is that the scattering rate at a few eV above the Fermi level is almost fully governed by the d-band density of states. This model also provides a useful guide for the analysis of possible experiments involving polarized electrons. For instance, it is known that some $Fe_{1-x}V_x$ alloys have a larger density of states at the Fermi level for the minority states, which inverts the magnetoresistance;[12] on the contrary, we see that this is of no importance in the case of electrons transmitted above the vacuum level.

I am deeply indebted to Catherine Bouton, A. Filipe, G. Lampel, Y. Lassailly, and especially to J. Peretti for a very careful reading of the manuscript. I am extremely grateful to M. I. D'Yakonov and to C. Hermann for many fruitful discussions. I thank C. Cacho for pertinent remarks. I acknowledge the ''Délégation Générale pour l'Armement'' for support.

[1] J. J. Quinn, Phys. Rev. **126**, 1453 (1962).
[2] C. R. Crowell, W. G. Spitzer, L. E. Howarth, and E. E. LaBate, Phys. Rev. **127**, 2006 (1962).
[3] M. P. Seah and W. A. Dench, Surf. Interface Anal. **1**, 2 (1979).
[4] H. Kanter, Phys. Rev. B **1**, 522 (1970).
[5] G. Schönhense and H. C. Siegmann, Ann. Phys. (Leipzig) **2**, 465 (1993).
[6] D. P. Pappas, K.-P. Kämper, B. P. Miller, H. Hopster, D. E. Fowler, C. R. Brundle, A. C. Luntz, and Z.-X. Shen, Phys. Rev. Lett. **66**, 504 (1991).
[7] D. R. Penn, S. P. Apell, and S. M. Girvin, Phys. Rev. B **32**, 7753 (1985).
[8] H.-J. Drouhin, A. J. van der Sluijs, Y. Lassailly, and G. Lampel, J. Appl. Phys. **79**, 4734 (1996).
[9] M. Getzlaff, J. Bansmann, and G. Schönhense, Solid State Commun. **87**, 467 (1993).
[10] A. Filipe, H.-J. Drouhin, G. Lampel, Y. Lassailly, J. Nagle, J. Peretti, V. I. Safarov, and A. Schuhl (unpublished).
[11] B. A. Gurney, V. S. Speriosu, J.-P. Nozières, H. Lefakis, D. R. Wilhoit, and O. U. Need, Phys. Rev. Lett. **71**, 4023 (1993).
[12] J.-P. Renard, P. Bruno, R. Mégy, B. Bartenlian, P. Beauvillain, C. Chappert, C. Dupas, E. Kolb, M. Mulloy, P. Veillet, and E. Vélu, Phys. Rev. B **51**, 12 821 (1995).