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Surface and Interface Analysis

Electron inelastic mean free paths: influence of the modelling energy-loss function

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Accurate quantitative surface analysis using electron spectroscopies is of paramount importance in nanoscale-based technology of today. In this context, an appropriate description of the electron inelastic mean free paths (IMFP) is mandatory. We have studied the influence of several model dielectric descriptions of the momentum-dependent energy-loss function (ELF), beyond the extensively used Penn's approach, on the calculation IMFP in the 200–2000 eV kinetic energy range. It is found that the theoretical description of the ELF based on the extended Mermin oscillators predicts systematically lower values for IMFP than those obtained by Penn's approach. The deviations are of 6–8% in the 1000–2000 eV energy range and increase up to 10–15% at 200–400 eV, depending upon the particular material and kinetic energy considered. A phenomenological expression is proposed to correct this effect. Copyright © 2007 John Wiley & Sons, Ltd.

KEYWORDS: electron inelastic mean free path; energy-loss function; electron spectroscopies

INTRODUCTION

Quantitative surface analysis using photoelectron or Auger electron spectroscopies is affected by a number of factors, among them the inelastic scattering during electron transport. This phenomenon is taken into account for quantification purposes through the differential cross section for inelastic scattering and the electron inelastic mean free path (IMFP). Both are closely related to the dielectric description of a material through the so-called electron energy-loss function (ELF). The ELF is defined as the imaginary part of a negative reciprocal complex dielectric function $\varepsilon(k,\omega)$, $\mathrm{Im}\{-1/\varepsilon(k,\omega)\}$, which describes the response of the medium to energy ω and momentum $\hbar k$ transfers. The IMFP for an electron travelling in an infinite medium is obtained from the ELF by the following expression:[1–3]

$$IMFP(E) = \left[\int_0^\infty \frac{d\omega}{\pi a_0 E}\int \frac{dk}{k} Im\left\{\frac{-1}{\varepsilon(k,\omega)}\right\}\right]^{-1} \qquad (1)$$

where a_0 is the Bohr radius and E the kinetic energy of the electron. In order to use the previous formula to calculate the IMFP, models for the momentum dependence of the ELF are needed. The most extensively and commonly used source of IMFP is the so-called TPP formulae.[4] They are based on calculations using Penn's statistical approach for the momentum-dependent ELF.[3] In his work, Penn proposed to describe the ELF as

$$\mathrm{Im}\left\{\frac{-1}{\varepsilon(k,\omega)}\right\}_{\mathrm{Penn}} = \frac{\omega - a(k)}{\omega}\mathrm{Im}\left\{\frac{-1}{\varepsilon(\omega - a(k))}\right\} \qquad (2)$$

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where $\varepsilon(\omega')$ is the optically measured (i.e. for $k = 0$) dielectric function and $a(k)$ is the dispersion term that, in a simplified form,[5–8] can be considered as

$$a(k) = \frac{\hbar^2 k^2}{2m} \qquad (3)$$

where m is the electron mass.

Factors affecting the accuracy of the determination of IMFP have been extensively discussed in the literature.[9] Thus, uncertainties related to the optical data, and those associated with the IMFP algorithms (ELF modelling, incorporation of surface effects) influence the evaluation of IMFP. Among them, the effect of the models describing the momentum-dependent ELF has not yet been considered in a systematic way. Besides Penn's approach described above, there are other descriptions of the dielectric behaviour of a material available in the literature. Ashley *et al*.[10] proposed an extended Drude oscillator and Abril *et al*.[11,12] used Mermin oscillators. Despite the great importance of the appropriate description of IMFP, it is not clear which one is most adequate, nor which is the influence of each theoretical description of the momentum dependence of the ELF in the determination of the IMFP.

In this paper, we first summarize the physical basis of the theoretical models to describe ELF (including momentum dependence) of materials with complicated electronic structure, i.e. beyond the free electron model. Then we show the influence of using various dielectric models on the determination of the IMFP.

THEORETICAL MODELS FOR THE ENERGY-LOSS FUNCTION

In this section, we briefly summarize the available models in the literature for real materials (beyond free electron-like materials).

Ashley *et al*.[10] proposed the simplest momentum-dependent ELF for real materials. It can be considered as an extended Drude model, consisting of the sum of terms as

$$\mathrm{Im}\left\{\frac{-1}{\varepsilon(k,\omega)}\right\}_{\mathrm{Drude}} = \sum_i \frac{A_i}{(\omega_{oi})^2}\mathrm{Im}\left\{\frac{-1}{\varepsilon_D(k,\omega)}\right\}_i \qquad (4)$$

Each so-called Drude oscillator takes the analytical form

$$\mathrm{Im}\left\{\frac{-1}{\varepsilon_D(k,\omega)}\right\}_i = \frac{(\omega_{oi})^2\gamma_i\omega}{[(\omega_{0i}+a(k))^2-\omega^2]^2+(\gamma_i\omega)^2} \qquad (5)$$

where A_i (eV^2), ω_{0i} (eV), γ_i (eV) represent the intensity, position and width of the oscillator. The dispersion term $a(k)$ is given in Eqn (3). This model emphasizes the influence of single particle interactions for large momentum transfer and interactions with plasmons for small momentum transfer. Each Drude term is a simplification of the Lindhard dielectric function.[1] This approach has been extensively used by many authors.[13–16] It is used to determine the inelastic scattering cross sections of electrons travelling close to surfaces as in the QUEELS software.[16]

Regarding the momentum dispersion considered in Penn's and extended Drude formalisms described in Eqn (3), it is worth mentioning that it has the same limit for large k than the complete expression deduced by Penn.[3] However, this simplified expression for $a(k)$ has stronger momentum dispersion for small k transferred. Other theories are also available for low k momentum transfer as the Random Phase Approximation (RPA).[17–19] RPA also predicts a weaker dispersion than as in Eqn (3) in the low k limit for free electron-like materials, such as Na, K, Al or Si. Besides, in experiments,[19] a weaker dispersion (than that predicted by the RPA theory) or even a negative dispersion in Cs is reported.[19] This strong material dependence of the dispersion may also have an influence on the determination of IMFP but we will not consider this effect in this paper. Nevertheless, it has been reported that Eqn (3) is expected to be a reasonable approximation to calculate IMFP.[7,8]

The momentum-dependent ELF proposed by Abril *et al*.[11,12] can be considered as an extended Mermin model. In this case, the ELF is expressed as a sum of oscillators:[20]

$$\mathrm{Im}\left\{\frac{-1}{\varepsilon(k,\omega)}\right\}_{\mathrm{Mermin}} = \sum_i \frac{A_i}{(\omega_{oi})^2}\mathrm{Im}\left\{\frac{-1}{\varepsilon_M(k,\omega)}\right\}_i \qquad (6)$$

Here, ε_M is the dielectric function derived by Mermin for a single oscillator. We refer to Refs. 11, 12, 20 to see the involved algebraic expression of each ε_M oscillator. The improvement of the Mermin description is that it takes into account the lifetime broadening of the excitations on momentum transfer and that this description is more robust for fulfilling sum rules for $k \neq 0$. It is worth mentioning that this model has been successfully used to calculate the stopping power of ions travelling in matter.[12]

Both Mermin and Drude oscillators converge on the same values for zero momentum (optical limit), i.e.

$$\mathrm{Im}\left\{\frac{-1}{\varepsilon(0,\omega)}\right\}_{\mathrm{Mermin}} = \mathrm{Im}\left\{\frac{-1}{\varepsilon(0,\omega)}\right\}_{\mathrm{Drude}} = \mathrm{Im}\left\{\frac{-1}{\varepsilon(\omega)}\right\}$$

Besides, it is worth noticing that the same set of parameters is able to describe the ELF either as Drude or Mermin expansions, which facilitates the comparison of the models.

Penn proposed to describe the ELF as Eqn. (2). Note that there is no need of parameterization of the ELF in this approximation to obtain the momentum and energy dependent ELF. However, it is straightforward to do it using an expansion of the ELF for $k = 0$ Å^{-1}

$$\mathrm{Im}\left\{\frac{-1}{\varepsilon(k,\omega)}\right\}_{\mathrm{Penn}} = \sum_i \frac{\omega-a(k)}{\omega}\mathrm{Im}\left\{\frac{-1}{\varepsilon\,(\omega-a(k))}\right\}_i \qquad (7)$$

where

$$\mathrm{Im}\left\{\frac{-1}{\varepsilon(\omega')}\right\}_i = \frac{A_i\gamma_i\omega'}{\left[{\omega_{0i}}^2-\omega'^2\right]^2+\left(\gamma_i\omega'\right)^2} \qquad (8)$$

This is convenient because it allows the same parameterization to be used for these three approximations for the momentum-dependent ELF.

Figure 1 shows the ELF calculated for Si, Ti, and Ag materials by the three models described above. The parameters used for each material are obtained from Ref. 13. Note that for $k = 0$ Å^{-1} the three models coincide as expected. Besides they deviate very little from the zero momentum transfer result and give similar results for $k < 1.0$ Å^{-1}. On the other hand, extended Drude and Penn models retain the initial structure for high momentum transfers (they behave very similarly in all the ranges of k tested) while in the case of extended Mermin it smeared out for k larger than 1.5–2.0 Å^{-1}. This latter behaviour coincides with the available experimental determination of ELF at $k \neq 0$,[21–23] and is not reproduced by either the Penn or extended Drude models.

This strong dependence on the modelling ELF for large momentum transfer may have an effect on the determination of IMFP. Therefore, it is mandatory to investigate and quantify this influence.

ELECTRON INELASTIC MEAN FREE PATHS

The main objective of this paper is to investigate the influence of the theoretical model for the ELF on the calculated IMFP. As mentioned above, using the extended Mermin model for describing response of the media to a perturbation, we expect a better description of the IMFP, because this model is based on more robust theoretical arguments[12,20] and reproduces experimental findings.[21–23]

We have performed a systematic evaluation of IMFP according to Eqn (1) by using the three-model ELF described above for a large set of materials (Al, Be, Cu, Si, Ti, Ag, Au). In order to quantify the deviation in the calculation of IMFP from Penn's description of the ELF, using the extended Drude or Mermin models, we have defined $\Delta_{\mathrm{Drude/Mermin}}$ as follows:

$$\Delta_{\mathrm{Drude/Mermin}} = 100^* \frac{\mathrm{IMFP}_{\mathrm{Drude/Mermin}} - \mathrm{IMFP}_{\mathrm{Penn}}}{\mathrm{IMFP}_{\mathrm{Penn}}} \qquad (9)$$

Thus, $\Delta_{\mathrm{Drude/Mermin}}$ represents the percentage deviation of the calculated IMFPs from the most frequently used

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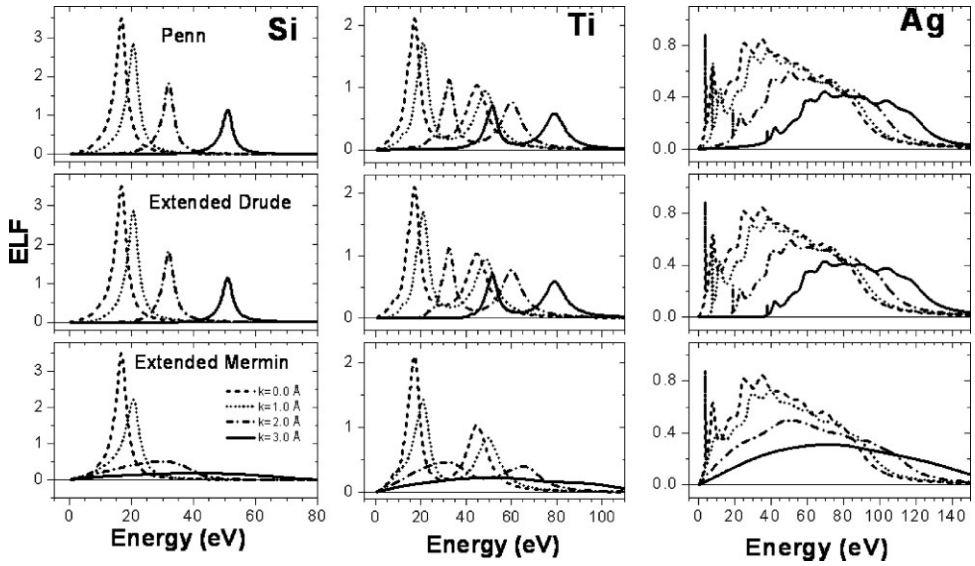

Figure 1. Momentum-dependent ELF corresponding to Si (left), Ti (middle) and Ag (right) calculated by Penn's approach (top) and extended Drude (middle) and extended Mermin (bottom) expansions, for $k = 0, 1, 2, 3$ Å^{-1}.

Penn's approach ($IMFP_{Penn}$), using either the extended Drude ($IMFP_{Drude}$) or Mermin ($IMFP_{Mermin}$) models described above. Results for several materials (Si, Ti, Ag and Au) are included in Fig. 2. We observe that the evaluation of IMFP is affected by the theoretical model used for the momentum dependent ELF. The Penn and extended Drude formalisms give similar results within 1–4% at high kinetic energies and within 2–6% at low kinetic energies. More significant deviations are observed when the extended Mermin model is considered. It gives 6–15% lower values than those obtained with the Penn description. The deviations are of 6–8% in the 1000–2000 eV energy range and increase upto 10–15% at 200–400 eV, depending on the particular material and the kinetic energy considered.

It is worth mentioning that we did not expect a huge difference in the IMFP determined by each model because small k values are the most important in the evaluation of IMFP, as it can be inferred by the dependence of the integral in Eqn (1) on $1/k$. However, it is important to note that the deviations depend on the kinetic energy of the electrons, and therefore, this will have consequences in concerning the accuracy of the quantitative analysis.

The results described above can be considered as systematic deviations of the IMFP obtained from TPP empirical predictive formula.[4] In order to supply a phenomenological predicted formula that reproduces the observed deviations of the $IMFP_{Mermin}$ from the IMFP obtained under Penn's approach ($IMFP_{Penn}$), Fig. 3 shows the correlation for several materials (Al, Be, Cu, Si, Ti, Ag, Au) and kinetic energies (200–2000 eV energy range) between the two different approaches for the IMFP. It is found that a linear regression reproduces rather well the difference between them, disregarding the type of material and kinetic energy, so that it can be expressed as follows:

$$IMFP_{Mermin} \approx -0.31 + 0.95 \times IMFP_{Penn} \qquad (10)$$

Equation (10) can be considered as a phenomenological way to correct $IMFP_{Penn}$ for incorporation of the momentum

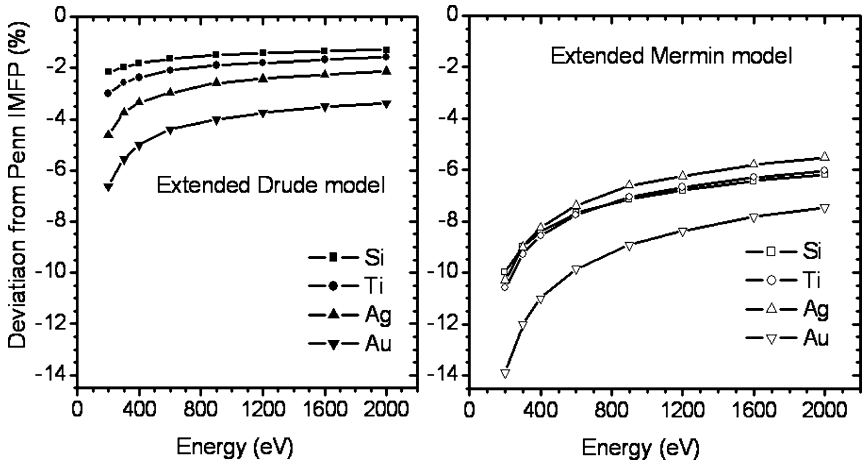

Figure 2. Left: Percentage deviation to calculate IMFP of several materials (Si, Ti, Ag and Au) from Penn's approach using the extended Drude model. Right: Percentage deviation to calculate IMFP of several materials (Si, Ti, Ag and Au) from Penn's approach using the extended Mermin model.

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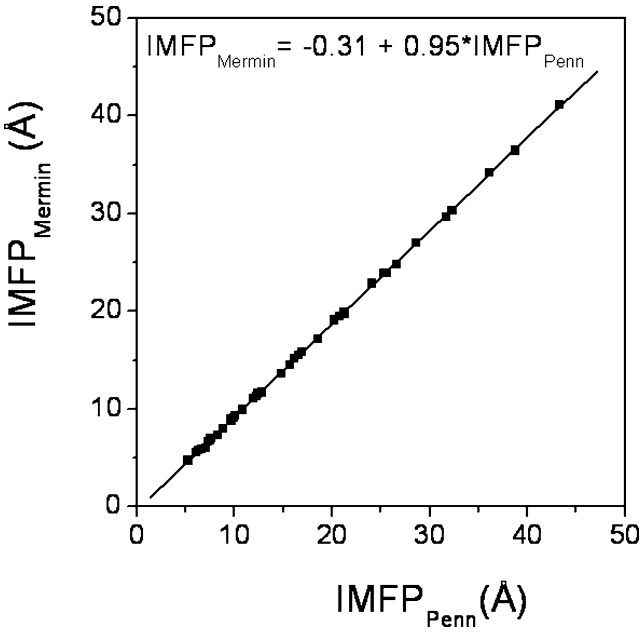

Figure 3. Correlation between the $IMFP_{Penn}$ and $IMFP_{Mermin}$ (in text) disregarding material type and kinetic energy of the electrons.

dependence of the ELF according to Mermin description. Note that for large values of IMFP, the deviation between the two approaches is 5% and that this deviation increases for smaller values of IMFP, as it was shown previously.

CONCLUDING REMARKS

We have shown that the choice of the model description of the momentum-dependent ELF affects significantly the evaluation of electron IMFP in the 200–2000 eV energy range, calculated according to Eqn (1). Thus, it is found that IMFP calculated using the most reliable model momentum-dependent ELF[12,21–23] (i.e. using extended Mermin oscillators), are systematically lower (5–16%) than those predicted by Penn's approach. The effect is stronger at lower kinetic energies and depends on each material considered. The IMFP value derived using Penn's approach can be considered as the upper limit for this magnitude. Taking into account that there are well established extensively used sources of IMFP (as TPP formulae[4]) based on Penn's approach, we have proposed a phenomenological way, according to Eqn (10), to correct for this effect.

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