Vacuum 136 (2017) 146–155

Contents lists available at ScienceDirect

Vacuum

journal homepage: www.elsevier.com/locate/vacuum

Differential inverse inelastic mean free path and differential surface excitation probability retrieval from electron energy loss spectra

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ARTICLE INFO

Article history:
Received 26 August 2016
Received in revised form
14 October 2016
Accepted 16 October 2016
Available online 19 October 2016

Keywords:
DIIMFP extraction
REELS deconvolution
PES
Invariant imbedding
XPS
Electron cross-section

ABSTRACT

Quantitative interpretation of the electron spectroscopy data requires the information on differential inverse inelastic mean free paths (DIIMFP) and differential surface excitation probabilities (DSEP). In this paper, we test an algorithm of extracting DIIMFP and DSEP from reflected electron energy loss spectra (REELS) and photo-electron spectra (PES) in which the desired functions are parametrized on the base of a classical Lorentz oscillator. Unknown parameters are found by using the fitting procedure. To account for surface excitations, the investigated samples are considered as multi-layer systems. Simulations of REELS and PES are performed by making use of the partial intensity approach. The partial intensities for the reflection function and the photo-electron density flux are computed on the base of the invariant imbedding method. Extracted DIIMFPs and DSEPs are compared with those obtained by other authors. Finally, REELS and PES spectra for Be, Mg, Al, Si, Nb and W are computed using the retrieved DIIMFPs and DSEPs, and compared with the experimental spectra. All comparisons show good agreement.

1. Introduction

Knowledge of the inelastic scattering parameters of solids is important for quantitative understanding of the energy loss process. The differential inverse inelastic mean free path (DIIMFP) and the differential surface excitation probability (DSEP) give the distribution of energy losses per unit path length in an individual inelastic collision in bulk and surface layers of solids, respectively. They are the main quantities characterizing inelastic scattering in solids. However, in practice, only integral quantities such as the inelastic mean free path (IMFP) and the electron stopping power are available in spectroscopic databases (e.g. NIST database by Ref. [1]). Studies involving linear response theory [2] can predict only the general shape of the DIIMFP, while using more sophisticated approaches (e.g. based on density functional theory [3]) is complicated for real atomic structures. Bearing that in mind, it seems to be more feasible to *extract* information on the DIIMFP and DSEP from experimental optical data [4] or REELS spectra rather than to compute them from basic physical principles.

A convenient numerical framework for the REELS spectra analysis is the partial intensity approach [5,6], in which a REELS spectrum is given by the weighted sum of multiple cross convolutions of DIIMFP and DSEP functions. The corresponding weighting factors are referred to as "partial intensities". To obtain DIIMFP and DSEP from REELS, the latter has to be *deconvolved* to filter out multiple scattering effects. Several techniques have been proposed to retrieve DIIMFP and DSEP from REELS spectra employing the partial intensity approach. A direct numerical inversion scheme was proposed by Ref. [7], and [8] to extract DIIMFP from a REELS spectrum. The P_1-approximation was used to compute the partial intensities. The main drawback of this scheme is that it does not take into account surface excitations. As a result, the extracted "effective" DIIMFP (being some kind of mixture of the actual DIIMFP, DSEP and their cross-convolution) can have negative values in the region corresponded to the cross-convolution of DIIMFP and DSEP. Further, this approach was extended by Werner (e.g., see Refs. [9,10]) to two-layer systems. There, DIIMFP and DSEP are retrieved from a pair of REELS spectra by reversing the bi-variate power series in Fourier space. A similar technique has been proposed by Ref. [11] which employs the REELS expansion through partial intensities only in the original space.

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http://dx.doi.org/10.1016/j.vacuum.2016.10.021
0042-207X/

Note, that all direct numerical inversion schemes above are severely ill-posed, i.e. the noise in the spectrum results in physically irrelevant peaks in the shapes of extracted DIIMFP and DSEP. To regularize the inversion [12], proposed to fit extracted functions to the Drude-Lindhard model and in this way to get a physically-consistent result.

The intent of this paper is to test a method for DIIMFP and DSEP retrieval from REELS and PES spectra in which the desired functions are parametrized on the base of the classical Lorentz oscillator. Unknown parameters of the model are found by means of the fitting procedure. An important part of our retrieval algorithm is the fast yet accurate method for partial intensity computations. It employs ideas of [13,14], and [15], and relies on the numerical solution of the invariant imbedding equations for scattered electrons. The forward simulations for REELS and PES are performed in the same framework, so that the DIIMFP functions extracted from REELS and PES can be cross-validated.

The rest of the paper is organized as follows. In Section 2, we briefly review the partial intensities approach and describe a technique for computing partial intensities using the invariant imbedding method. Section 3 provides basic relations for reflection and transmission functions in the case of multi-layer systems. A description of the retrieval algorithm set-up follows in Section 4. Here, DIIMFP and DSEP functions are extracted from REELS and PES spectra for a set of materials (Be, Mg, Al, Si, Nb and W). Section 5 summarizes the present work and outlines future tasks.

2. Evaluation of partial intensities for single layers

In this section we consider a single layer illuminated by the electron beam or the X-ray irradiation. Let $R(\tau,\Delta,\mu_0,\mu,\varphi)$ be the reflection function of electrons, i.e. the ratio of the outgoing flux per unit solid angle per unit energy interval to the incident flux [16], $\tau=z/l_{tot}$ is the dimensionless layer thickness, z is the geometrical thickness of the layer, $l_{tot}=[n(\sigma_{in}+\sigma_{el})]^{-1}$ is the total mean free path, n is the concentration of scatters, σ_{el} and σ_{in} are the elastic and inelastic scattering cross-sections, respectively, Δ is the energy loss, μ_0 is the cosine of the incident polar angle θ_0, μ is the cosine of the viewing polar angle θ, φ is the azimuthal angle between incident and sighting directions, as shown in Fig. 1. Expanding the reflection function into a Fourier cosine series gives

$$R(\tau,\Delta,\mu_0,\mu,\varphi)=\sum_{m=0}^{\infty}(2-\delta_{m0})R^m(\tau,\Delta,\mu_0,\mu)\cos(m\varphi). \quad (1)$$

Here $\delta_{mm'}$ is the Kronecker delta. Within the partial intensity approach, the functions R^m can be expanded as follows:

$$R^m(\tau,\Delta,\mu_0,\mu)=\sum_{k=0}^{\infty}R_k^m(\tau,\mu_0,\mu)x_{in}^k(\Delta), \quad (2)$$

where $x_{in}^0(\Delta)=\delta(\Delta)$ is the Dirac function, $x_{in}^1(\Delta)=x_{in}(\Delta)$ is the probability distribution of the energy loss in a single inelastic event (also referred to as normalized DIIMFP or NDIIMFP), and $x_{in}^k(\Delta)$ is the spectrum of energy losses after k successive inelastic scattering events. The latter is computed as the k-fold self-convolution:

$$x_{in}^k(\Delta)=\int_0^{\Delta}x_{in}^{k-1}(\varepsilon)x_{in}(\Delta-\varepsilon)d\varepsilon.$$

In practice, the summation in Eq. (2) is performed up to the K-th term, where K is the maximum number of inelastic scattering collisions taken into account. The transmission function $T(\tau,\Delta,\mu_0,\mu,\varphi)$ and the photo-electron flux density $Q(\tau,\Delta,\mu_0,\mu,\varphi)$ are expanded analogously as in Eqs. (1) and (2) providing the partial intensities $T_k^m(\tau,\mu_0,\mu)$ and $Q_k^m(\tau,\mu_0,\mu)$. To simplify notations, hereafter we suppress the m-superscript with the dependence on azimuthal mode assumed. Note, that $R_k(\tau,\mu_0,\mu)$, $T_k(\tau,\mu_0,\mu)$ and $Q_k(\tau,\mu_0,\mu)$ refer to the k-fold inelastically scattered particles, while $R_0(\tau,\mu_0,\mu)$, $T_0(\tau,\mu_0,\mu)$ and $Q_0(\tau,\mu_0,\mu)$ refer to the elastically scattered electrons.

Assuming the Poisson stochastic process for multiple energy losses [17], the energy distribution of electrons with the path length τ is written as

$$L(\tau,\Delta)=\sum_{k=0}^{\infty}L_k(\tau,\Delta)=\sum_{k=0}^{\infty}\left\{\exp(-\tau)\frac{(1-\lambda)^k\tau^k}{k!}x_{in}^k(\Delta)\right\}, \quad (3)$$

where λ is the single scattering albedo, and $L_k(\tau,\Delta)$ is the distribution of energy losses after k-fold scattering as a function of τ [18,19].

To compute partial intensities we adopt the concept of invariant imbedding, which is due to [20]. He derived an equation for reflection from a semi-infinite atmosphere by noting that the reflection function remains unchanged upon addition of a new layer. This technique was generalized by Ref. [21] for a finite layer. The extension of this method to the partial intensity approach is described in Ref. [22]. Derivation of equations for functions R_k, T_k, and Q_k involves the following steps [23]:

1 add an infinitely thin layer to the layer;
2 consider single scattering processes in that layer which contribute to the change in R_k, T_k, and Q_k;
3 express R_k, T_k, and Q_k functions for the system "sample + layer" through corresponding functions for the sample.

The resulting equations for elastically scattered electrons (k=0) read as follows:

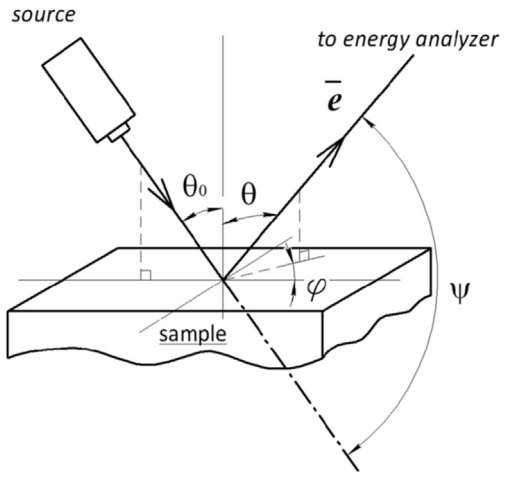

Fig. 1. Illustration of the experimental geometry. A sample is illuminated by the electron beam or the X-ray irradiation. Here θ_0 is the polar angle of incidence, θ is the viewing polar angle, φ is the azimuthal angle between incident and sighting directions and ψ is the scattering angle.