

Review (Invited)

# Modeling of Electron Transport in the Surface Region of Solids: Metrology of Quantitative Analysis by Electron Spectroscopies

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Quantification of XPS and AES analyses requires a theoretical model that relates measured signal intensity with a given quantity describing a studied surface region. Determination of the following quantities and parameters facilitating quantification is addressed here: surface composition, overlayer thickness, and sampling depth of a particular measurement. It is shown that parameterization of quantitative analysis by XPS and AES can be based on the emission depth distribution function for the signal electrons. From this function, the formalism for numerous quantitative applications can be derived. Reliable sources of relevant parameters are briefly discussed.

## 1. Introduction

Quantification of surface-sensitive electron spectroscopies is founded on theoretical models that describe transport of signal electrons in the surface region of solids. These models for typical analytical applications can be systematized into two major groups: (a) transport of signal electrons emitted at a certain depth by X-ray radiation or by a high-energy electron beam; (b) transport of electron elastically backscattered from a solid surface. Much material concerning the experimental and theoretical aspects of quantitative analysis has been published. An interested reader is referred to monographs summarizing these issues (see e.g., refs [1-3]).

A formalism that relates a needed quantity characterizing the surface region with measured spectrum features can be very complicated due to multiple elastic and inelastic interactions of the signal electrons. Thus, to predict electron trajectories in solids, we need to have an accurate and reliable mathematical formalism for the individual scattering events. Monte Carlo simulations implementing different strategies can be universally used for modeling multiple interactions. Since this approach requires a considerable computer effort, it is impractical

in routine calculations. Different simplified theoretical models of various degrees of accuracy are typically used. Attention should be drawn to transport theory involving the Chandrasekhar function [4,5] which has an accuracy comparable to the Monte Carlo simulation algorithms, yet the relevant calculations are considerably faster. This formalism can be readily extended for photoelectron energies of interest in high-energy photoelectron spectroscopy (HAXPES) [5].

Computational algorithms used for practical applications are facilitated if we use simplified theoretical models describing electron transport in solids. For example, elastic electron-scattering effects are frequently neglected in calculations although we know that they may considerably affect the calculated signal intensity. To perform calculations using more realistic theoretical models, additional correction parameters are introduced that make possible the use of a simplified analytical formalism, yet the accuracy of calculations is considerably improved. For convenience of practical analysis, these parameters should be readily available for a given electron energy, analyzed solid, and experimental configuration. This issue is briefly overviewed in the present report.

**2. Theory**

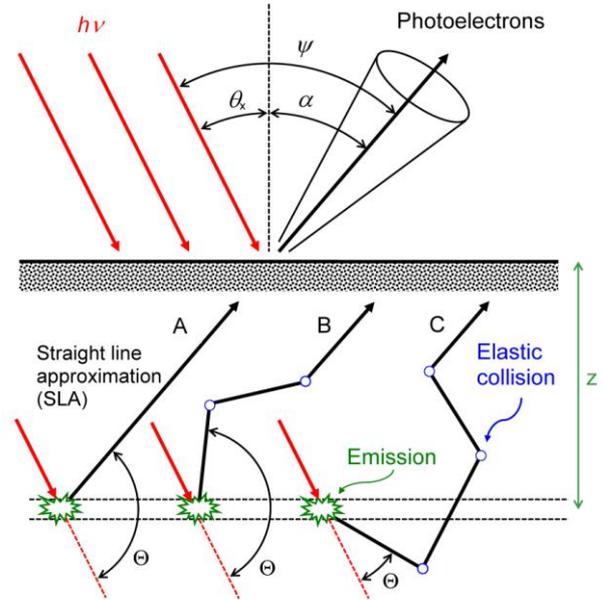
Let us consider photoelectron trajectories emitted at a certain depth,  $z$ . These trajectories, in particular trajectories of electrons entering an analyzer, are governed by the direction of electron emission in the sample,  $\Theta$ , and later direction changes due to elastic-scattering events. This is illustrated in Fig. 1. Generally, we can distinguish three groups of electron trajectories: electrons emitted towards the acceptance angle of the analyzer which do not undergo elastic interactions (group A), electrons emitted towards the surface that enter an analyzer due to elastic collisions (group B), and electrons emitted towards the bulk of the solid (group C).

We will consider here parameterization of three quantitative applications of electron spectroscopies: (i) determination of surface composition, (ii) overlayer thickness measurements, and (iii) estimation of sampling depth of a particular measurement. Theoretical aspects justifying different parameterization are extensively analyzed in the literature (see e.g., Tanuma in ref. [2], pp 259-294, Jablonski and Powell [6]). To ensure comparability of parameters determined in different laboratories, their precise definitions are standardized [7].

It has been shown [5,6] that the basic parameter that describes signal-electron transport in a solid is the emission depth distribution function (EMDDF),  $\phi(z, \alpha)$ . It is defined as the “probability that the particle or radiation leaving the surface in a specified state and in a given direction originated from a specified depth measured normally from the surface into the material” (ref. [7], definition 4.161). The EMDDF derived for a sample with planar surface on the assumption that elastic-scattering effects can be neglected (trajectories designated by A in Fig. 1) is expressed by a simple equation

$$\phi^{SLA}(z, \alpha) = \frac{d\sigma_x}{d\Omega} \exp\left(-\frac{z}{\lambda_{in} \cos \alpha}\right) \dots\dots\dots(1)$$

where  $d\sigma_x/d\Omega$  is the photoemission cross section (PCS), and  $\lambda_{in}$  is the inelastic mean free path (IMFP). The considered theoretical model is frequently called the straight-line approximation (SLA). To determine a more accurate EMDDF,  $\phi(z, \alpha)$ , we have to account for the elastic-scattering events. For the relevant Monte Carlo simulations, we need to know the differential elastic-scattering cross section (DCS),  $d\sigma_{el}/d\Omega$ , from which the probability density function of scattering an-



**Fig. 1.** (Color Online) Outline of photoelectron emission from a given layer in the surface region, and the notation used.

gles can be derived. On the other hand, analytical expressions for the EMDDF derived from transport theory require knowledge of a related quantity, i.e. the transport mean free path.

$$\lambda_{tr} = \left(2\pi N \int_0^\pi (1 - \cos \theta_{el}) \frac{d\sigma_{el}}{d\Omega} \sin \theta_{el} d\theta_{el}\right)^{-1} \dots\dots\dots(2)$$

where  $N$  is the atomic density (number of atoms per unit volume), and the integration is extended over all scattering angles,  $\theta_{el}$ . We show below that numerous parameters used for quantification can be expressed by the EMDDF.

**2.1. The photoelectron signal intensity**

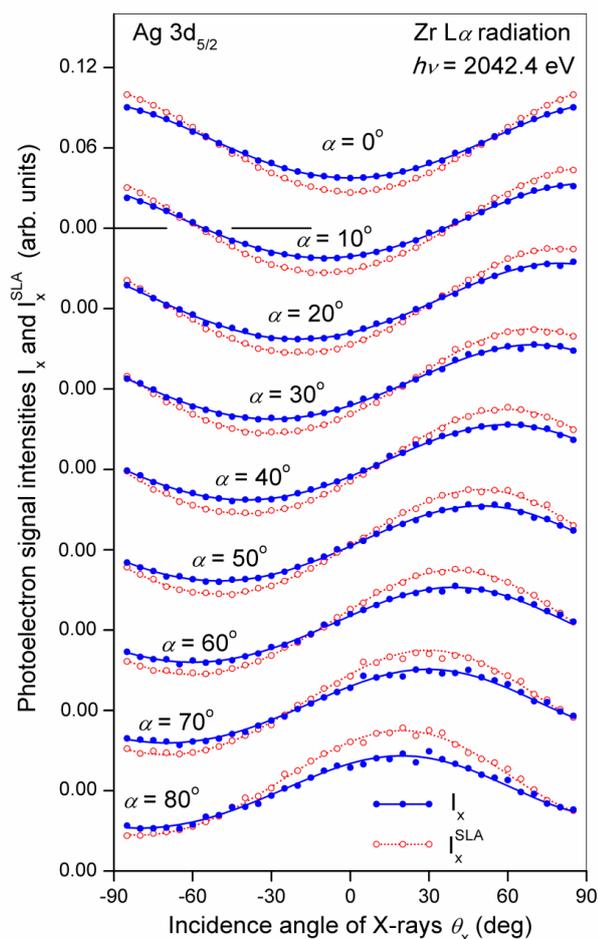
The recorded photoelectron (or Auger electron) signal intensity is related to the EMDDF by the following simple expression

$$I_x \propto N \int_0^\infty \phi(z, \alpha) dz \dots\dots\dots(3)$$

For the SLA model, as follows from Eq. (1), the signal intensity is given by

$$I_x^{SLA} \propto \frac{d\sigma_x}{d\Omega} \lambda_{in} N \cos \alpha \dots\dots\dots(4)$$

To simplify the formalism, all instrumental parameters in Eqs (3) and (4) are omitted. The influence of elas-



**Fig. 2.** (Color Online) Photoelectron signal intensities due to Ag 3d<sub>5/2</sub> photoelectrons excited by Zr L $\alpha$  X-rays as a function of geometry defined by angles  $\theta_x$  and  $\alpha$  (see Fig. 1). Open circles and dotted line: straight-line approximation (SLA); filled circles and solid line: Monte Carlo strategy taking into account photoelectron elastic collisions.

tic-scattering effects on HAXPES signal intensity is known to depend on measurements conditions [8]. A comparison of Ag 3d<sub>5/2</sub> photoelectron intensities excited by Zr L $\alpha$  X-rays, calculated from two theoretical models, is shown in Fig 2. We see, that there are a number of experimental configurations defined by the angles  $\theta_x$  and  $\alpha$  in which the neglect of elastic electron interactions may considerably affect the signal intensities. It is thus recommended, for safety, to base the quantitative analysis on an accurate formalism that includes corrections for electron-scattering effects.

**2.2. Parameters expressing the sampling depth of electron spectroscopies**

The sampling depth for AES and XPS is generally different from the IMFP and depends on the experi-

mental configuration. The relevant information is described by the mean escape depth (MED) and the information depth (ID). According to the ISO definition, the MED is: “... an average depth normal to the surface from which the specified particles or radiations escape“ (ref. [7], definition 4.203). This parameter is related to the EMDDF by

$$D = \frac{\int_0^\infty z \phi(z, \alpha) dz}{\int_0^\infty \phi(z, \alpha) dz} \dots\dots\dots(5)$$

The ID is defined as: “... a maximum depth normal to the surface from which useful information is obtained” (ref. [7]; definition 4.246). The ID,  $S$ , is related to the EMDDF by the equation

$$\frac{\int_0^S \phi(x, \alpha) dz}{\int_0^\infty \phi(x, \alpha) dz} = \frac{P}{100} \dots\dots\dots(6)$$

where  $P$  is the percentage of the photoelectron signal intensity due to the layer of thickness  $S$ . Typically, we assume that  $P = 95\%$  or  $99\%$ . On assumption of a particular percentage,  $P$ , Eq. (6) is generally a nonlinear equation with respect to  $S$ , and should be solved using numerical methods [5]. However, Eqs (5) and (6) considerably simplify for the SLA model. Introducing Eq. (1) into these equations, we obtain

$$D^{SLA} = \lambda_{in} \cos \alpha \dots\dots\dots(7)$$

$$S_p^{SLA} = -\lambda_{in} \cos \alpha \ln \left( 1 - \frac{P}{100} \right) \dots\dots\dots(8)$$

**2.3. Effective attenuation length**

The effective attenuation length (EAL) is a convenient term for taking elastic-scattering effects on signal intensities into account. It is: “... a parameter which, when introduced in place of the inelastic mean free path into an expression derived for AES and XPS on the assumption that elastic scattering effects are negligible for a given quantitative application, will correct that expression for elastic scattering effects” (ref. [7]; definition 4.35). In the terminology standard, the users of this parameter are warned that the EAL values may be different for different quantitative applications. Let us briefly address this issue below.

The most frequent use of the EAL is the determination of an overlayer thickness. For this measurement, the EAL derived for an overlayer deposited on a planar sub-

strate,  $L_{TH}$ , has the following form (see e.g., Tanuma in ref. [2], pp 259-294, Jablonski and Powell [6,9]):

$$L_{TH} = \frac{1}{\cos \alpha} \frac{t}{\left[ \ln \int_0^\infty \phi(z, \alpha) dz - \ln \int_t^\infty \phi(z, \alpha) dz \right]} \dots (9)$$

where  $t$  is an overlayer thickness. In experimental configurations of practical interest, the parameter  $L_{TH}$  is a weak function of  $\alpha$  and  $t$ , and can be averaged without minor loss in accuracy. Details of experimental procedures and formalism used for determination of an overlayer thickness are not provided here; an interested reader is referred to relevant literature (e.g., refs [1-3]).

Suppose that we need to determine the depth of a thin layer embedded in a matrix of a given material (a so-called marker-layer). The relevant formalism requires knowledge of another EAL,  $L_{ML}$ . This parameter is related to the EMDDF as follows [9]:

$$L_{ML} = \frac{1}{\cos \alpha} \frac{z_{ML}}{\left[ \ln \phi(0, \alpha) - \ln \phi(z_{ML}, \alpha) \right]} \dots (10)$$

where  $z_{ML}$  is the depth of the marker layer. Practical use of this parameter is possible for measurements conditions in which the EAL,  $L_{ML}$ , weakly depends on  $\alpha$  and the depth  $z_{ML}$  [9].

The sensitivity factor approach for determination of surface composition is based on measurements of peak intensities due to elements present in the surface region. To account for elastic-scattering effects, we need to replace the IMFP in the SLA formalism for each measured intensity with the following EAL (Tanuma in ref. [2], pp 259-294, Jablonski and Powell [9])

$$L_C = \lambda_{in} \frac{I_x}{I_x^{SLA}} = \lambda_{in} \frac{\int_0^\infty \phi(z, \alpha) dz}{\int_0^\infty \phi^{SLA}(z, \alpha) dz} \dots (11)$$

Eqs (9)-(11) are distinctly different, and thus we may expect that the numerical values of these EALs should generally differ. On the other hand, these EALs are identical for the SLA formalism

$$L_{TH}^{SLA} = L_{ML}^{SLA} = L_C^{SLA} = \lambda_{in} \dots (12)$$

### 2.4. Other parameters

In the formalism of quantitative AES, we need to determine a contribution due to elastic and inelastic scattering of primary electrons to the measured intensity. The

needed correction is called the backscattering correction factor (BCF). It is: "... the ratio of the total Auger-electron current arising from ionizations in the sample caused by both the primary electrons and the backscattered electrons to the Auger-electron current arising directly from the primary electrons" (ref. [7]; definition 7.3). The BCF is also related to the EMDDF:

$$R = \int_0^\infty \Phi(z, E_c, E_0) \phi(z, \alpha) dz \dots (13)$$

where  $\Phi(z, E_c, E_0)$  is the excitation depth distribution function that describes the density of ionizations of a given subshell,  $E_c$  is the threshold energy of ionization, and  $E_0$  is the primary-electron energy.

It has been shown that the concept of the EMDDF can be generalized by including the inelastic-scattering events in the electron trajectory towards an analyzer [10,11]. The relevant parameter, called the partial escape distribution (PED), is useful in calculations of the background in the spectra, and is defined as the probability distribution for the process in which an electron generated in a certain depth interval will escape from the surface with a direction in a certain angular interval after experiencing a certain number of inelastic scattering processes in the sample (see ref. [7], definition 4.323). For  $n$  inelastic interactions, the relation to the EMDDF is the following [4,11]:

$$P_n(z, \alpha) = (-1)^n \frac{\nu^n}{n!} \frac{d^n \phi(z, \alpha)}{d\nu^n} \dots (14)$$

where  $\nu = 1/\lambda_{in}$

### 3. Sources of parameters

Let us tentatively systematize the parameterization of quantitative application of AES and XPS. A preliminary proposition is outlined in Fig. 3. The diagram stresses the fact that the EMDDF is a generic parameter from which originate other parameters used in different quantitative applications. To calculate the EMDDF, we need to describe the elastic and inelastic interactions of the signal electrons. We need to know the IMFP to estimate the probability of energy loss for the signal electrons. The probability density function of elastic scattering angles can be derived from the DCSs

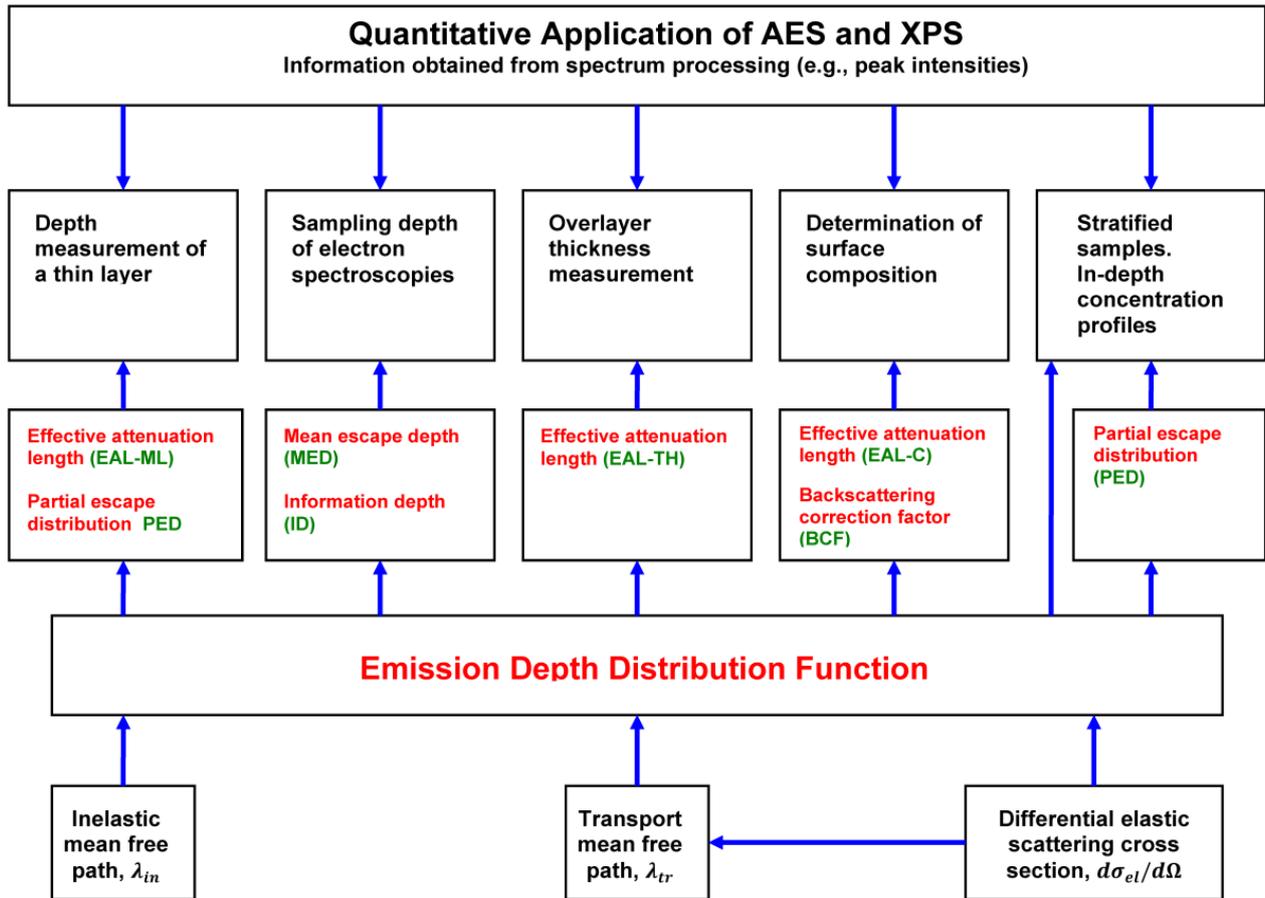


Fig. 3. (Color Online) Parameterization of selected quantitative applications of electron spectroscopies.

$$H(\theta_{el}) = \frac{2\pi}{\sigma_{el}} \frac{d\sigma_{el}}{d\Omega} \sin \theta_{el} \dots\dots\dots (15)$$

where  $\sigma_{el}$  is the total elastic-scattering cross section. In the analytical transport theory, the elastic-scattering effects in the expression for the EMDDF are accounted for with sufficient accuracy with one parameter, i.e., the transport mean free path [5,9]. Thus, sources of accurate values of IMFP and DCS for a given electron energy would be sufficient to derive numerous other parameters needed for quantification. However, the relevant formalism and the corresponding computational algorithm are typically very complicated (e.g. the expression for the EMDDF from transport theory [5,9]), and cannot be easily edited for a particular analytical problem. Consequently, different compilations of these parameters have been published. However, a convenient source of EMDDFs and related parameters would be a computer-controlled database that provides needed information in an electronic form. Such an effort has been made in

the Standard Reference Data Program run by the National Institute of Standards and Technology. In Table 1, information is listed on particular databases and parameters that are contained in these databases. The database content was carefully evaluated to make sure that the users obtain accurate data for quantification procedures.

**4. Concluding remarks**

The parameterizations described above are limited to quantification procedures involving parameters that can be derived from the emission depth distribution function. There are also other techniques based on theory of electron transport that can be supported by suitable parameterization. One can mention here elastic-peak electron spectroscopy (EPES) and its analytical applications. An important application of EPES is the determination of electron inelastic mean free paths. An accurate theoretical model is needed for calculations of the elastic electron-backscattering probability. With few exceptions, Monte Carlo simulations are used in the published re-

**Table 1.** Parameters compiled in selected NIST databases

Database	Parameters
NIST Electron Elastic-Scattering Cross-Section Database [12] <sup>a</sup>	$d\sigma_{ei}/d\Omega, \sigma_{ei}, \lambda_{tr}$
NIST Electron Inelastic-Mean-Free-Path Database [13]	$\lambda_{in}$
NIST Electron Effective-Attenuation-Length Database [14] <sup>b</sup>	$L_{TH}, L_{ML}, L_C, D, \phi(z, \alpha)$
NIST Database for the Simulation of Electron Spectra for Surface Analysis [15]	$d\bar{\sigma}_{ei}/d\Omega, \sigma_{ei}, \lambda_{tr}, \lambda_{in}, R$
NIST Backscattering-Correction-Factor Database for Auger Electron Spectroscopy [16]	$R, \phi(z, \alpha)$

<sup>a</sup> The database contains the transport cross sections [equal to the integral in Eq. (2)] from which the transport mean free paths can be readily calculated for a given solid.

<sup>b</sup> The attenuation lengths  $L_C$  are available only for Auger electron spectroscopy (parameter  $Q_A = L_C/\lambda_{in}$ ).

ports. However, an analytical theoretical approach of comparable accuracy was recently proposed for EPES analytical applications [17-19]. In the relevant analytical formalism, we need parameters which are coefficients of the expansion of the DCS in a series of Legendre polynomials.

Quantification of XPS measurements requires knowledge of the PCSs,  $d\sigma_x/d\Omega$  [see Eq. (1)]. They are needed for typically used laboratory sources: Mg  $K\alpha$   $h\nu = 1253.6$  eV and Al  $K\alpha$   $h\nu = 1486.6$  eV. However, extensive tabulations of these parameters published by Trzhaskovskaya et al. [20-22] contain data for all elements and photoelectron lines and for kinetic energies in the range up to 10 keV. Thus, the parameterization of quantitative XPS analysis for HAXPES applications can be extended for this energy range [5,23]. One should also mention that the formalism of XPS refers to excitation of the surface region by unpolarized radiation from laboratory X-ray sources or by circularly polarized synchrotron radiation. This formalism generally does not apply to excitations by polarized X-rays since the relevant theoretical model needs additional information on direction of the polarization vector with respect to the surface

normal. Detailed studies of this effect are planned for the future. An attempt to calculate the MEDs for photoelectrons emitted by the polarized radiation has recently been published by Tanuma et al. [24]. A powerful source of PCSs for kinetic energies exceeding 10 keV seems to be the theoretical model of Sabbatucci and Salvat [25]. The program PHOTACS edited by these authors, implementing their theory, is a universal source of PCSs for photoelectrons emitted by unpolarized and polarized X-rays of any energy reaching even 500 keV. Thus, edition of new complete databases of additional parameters would be very useful since high-quality physical data considerably facilitates software development and thus quantification for practical analysis.

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**Discussions and Q&A with Reviewers****Reviewer #1 Cedric Powell (NIST)**

The author provides a concise and useful report of a formalism for quantitative surface analysis by AES and XPS with emphasis on determinations of surface composition, overlayer thickness, and sampling depth. A number of needed parameters can be obtained from the emission depth distribution function, and the author provides information on sources of needed data. Figure 3 is a very useful pictorial summary of the approach. I recommend acceptance for publication subject to the author's response to the following minor comments.

**[Q1\_1]**

It seems undesirable in Ref. [12] to refer to an older version of the SRD 64 database. I recommend deletion of this reference.

**[A1\_1]**

I agree. I have corrected Ref. [12]. However, I added the web address to the older version indicating that it contains additional data that may be useful in some applications (phase shifts).

**[Q1\_2]**

The author provides a concise and useful summary of a formalism for quantitative surface analysis by AES and XPS. The XPS summary does not refer explicitly to possible X-ray sources. Will the author please comment on the range of X-ray energies for which the formalism is valid and provide a few more details (e.g., from [5]) on how the use of polarized X-rays from synchrotron sources might modify the results.

**[A1\_2]**

Information requested by the Referee is added in the last paragraph of Concluding Remarks. The energy ranges, and the applicability of the XPS formalism to surface excitation by laboratory X-ray sources or by circularly polarized synchrotron radiation is indicated. The Reader is warned that the presented formalism does not apply to photoelectron emission by polarized X-ray radiation.

**Reviewer #2 Shigeo Tanuma (NIMS)**

This paper well describes the modeling of electron transport in a surface region that is the basis of quantification of XPS and AES. This is very useful for most researchers and engineers who are interested in surface analysis by electron spectroscopy. Then, I recommend to publish it in JSA after reconsidering the following a point.

**[Q2\_1]**

Equation (11):

$$L_C = \lambda_{in} \frac{\int_0^{\infty} \phi(z, \alpha) dz}{\int_0^{\infty} \phi^{SLA}(z, \alpha) dz}$$

is unclear for most readers. Then, I would like to ask the author to describe more clearly. The reason is as follow. The  $\phi^{SLA}$  was given by Eqn (1). However,  $\phi$  was not given clearly. Then, it is hard to understand the Eqn (11). Then, you should provide more information about Eqn (11). For example, Eqn (11) can be described as

$$L_C = \lambda_{in} \frac{I_x}{I_x^{SLA}} = \lambda_{in} \frac{\int_0^{\infty} \phi(z, \alpha) dz}{\int_0^{\infty} \phi^{SLA}(z, \alpha) dz}$$

**[A2\_1]**

The EMDDF,  $\phi(z, \alpha)$ , is explained in the second sentence following Eq. (1). However, I agree that the modification suggested by the referee would clarify the definition of  $L_C$ . Eq. (11) is presently written in the suggested form.