Monte Carlo simulation of reflection electron energy loss spectra

Thesis of Ph. D.

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Introduction

Electron emission might be excited by particle (e.g. electron or photon) radiation. A possible way of the analysis of samples is to measure the energy of the emitted electrons of the sample. Surface sensitive electron spectroscopies, like *Auger Electron Spectroscopy* (*AES*), X-ray *Photoelectron Spectroscopy* (*XPS*) [I1] and *Reflection Electron Energy Loss Spectroscopy* (*REELS*) [I2] are based on this principle. A common feature of these methods that they collect signals in the energy range of some hundred *eV* from a given thickness of the sample.

Regarding *AES* as an example, we can illuminate, how surface sensitive electron spectroscopies work. *Auger* electrons loose energy during their way to surface, thus reducing their original intensity because of transport. Therefore the measured intensity depends not only on the number of excited atoms, but also on their spatial distribution. *Inelastic Mean Free Path* (*IMFP*), that is, the average path length between two succeeding inelastic events [I3] determines the frequency of energy losses of an *Auger* electron. The width of the investigated layer can be reduced by applying *Auger* electrons of lower energies, which automatically means lower *IMFP* values. For evaluating experimental data, knowledge of *IMFP* values is necessary. The most reliable experimental method that determines *IMFP* is *Elastic Peak Electron Spectroscopy* (*EPES*), which is introduced by György Gergely [I4].

Measurements of inhomogeneous samples with depth-dependent concentration are often combined with other methods. Materials are often sputtered with energetic ion beams during depth profiling, thus the concentration distribution in the surface region of the sample is changing continously.

AES, *XPS* and *EPES* depth profiling measurements [I5] can be carried out with this technique. To interpret the results of these types of measurements a description of electron transport is needed.

Several articles are known to describe the electron transport in homogeneous sample [I6]. In these descriptions amorphous, semi-infinite materials with constant density and without any defects are usually taken,. Further assumtions are made on a perfectly smooth surface of the samples, neglecting segregation and contamination. Electron transport is described by elastic and inelastic scattering events.

Reliable calculation methods of elastic scattering are known, and their numerical results are available from publications [I7]. Gergely and

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Jablonski determined *IMFP* data from *EPES* experiments first time considering multiple elastic scattering [I8].

Inelastic scattering can be described either by using local approximation or in the framework of dielectric theory. Experimental optical data provide the dielectric function, which result in *IMFP* data. Obviously this *IMFP* reflects the characterics of bulk dielectric function. The simplest description for the interaction of solid and electron is the jellium model [I9]. This model assumes non-binding (free) electrons moving in the potential field of uniformly distributed positive charges, neglecting the band stucture of real solid samples. Jellium model usually estimates *IMFP* values well [I10]. Tougaard suggested simple, easy-to-use formulae for the calculation of energy loss functions [I11]. Tougaard's method determines energy loss function from the measured spectra. However, his method neglects surface excitations.

Ritchie predicted the possible excitations of surface plasmons by electrons in 1957 [I12]. Analysis of *REELS* spectra provides a suitable way for investigating the properties of surface excitations. Károly Tőkési applied his three-layer model for investigating excitations [I13-I15]. This model devided the sample into bulk and surface regions separating them from the vacuum. Even the excitations of *Ag*, which have complicated energy loss structure, can be examined applying his method [I13-I14].

Position- and direction dependences of surface excitations had been known for a long time ago; Chen and Ding reported depth-dependent energy loss functions [I16-I17]. Although these detailed calculations result similar shapes, the actual data of the energy loss functions are different [I17].

Surface excitations can be described by the Surface Excittion Parameter (*SEP*), which is the average number of surface excitation while an electron is crossing the surface once [I18]. Simple, analytical formulae are available to calculate the total *SEP* values [I19-I20]. On the basis of Oswald's work [I21] Werner gave a formula and material parameters for some materials [I21]. Using this formula and material parameters, *SEP* values can be calculated for arbitrary electron energy and for any measurement geometry.

Evaluating measured results surface excitations should be considered. This can be made by Chen's or Ding's results assuming ideal samples. It is also important to note that the measured samples may considerably differ from the ideal ones. This means that for an actual sample many physical parameters are not available.

There are no reliable description of electron transport, which can be applied for evaluating measured data in an inhomogeneous enviroment.

Abstracts of Conferences

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4th **point**

I determined the *IMFP* values of SiO_2 and Si_3N_4 using *EPES* method with *Si* reference sample. I made surface correction on the measured elastic peak intensity of *Si*, using material parameters and formula of Chen. I showed that this surface correction is necessary.

5th **point**

I simulated *EPES* depth profiling measurements on *Ge-Si* multilayered samples for the geometries of *CMA*, *ESA 31* and *DESA 100* spectrometers. For each geometry I determined the optimum electron energy for the measurements. I evaluated differences in the calculated elastic peak profiles due to the different measurement geometry.

My goal is to develop a simple, efficient method to evaluate experimetal results of electron spectroscopies. In my PhD thesis I dealt with the evaluation of EPES and REELS spectra. I applied Monte Carlo method to describe the electron transport. I developed a Monte Carlo model that can be used to describe electron transport in multi-component, homogeneous and inhomogeneous (multilayered) samples. This Monte Carlo code was applied in evaluation of experimentally measured spectra.

Results

I simulated *EPES-REELS* experimental spectra (of *polySi*, *amorphous-Ge* and *microcrystalline Sn* samples [I25]) measured with a *DESA 100* [I24] electron spectrometer in MFA and with *ESA 31* electron spectrometer [I25] in ATOMKI. Elastic scattering was described by using elastic differential cross section data published by NIST [I26]. Description of inelastic scattering treated surface and volume excitations seperately, applying surface and volume energy loss functions. While the shapes (the energy distributions) of the volume energy loss function were constructed by the sum of Drude-type functions [I27], the shape of the surface energy loss function consisted of a single Drude-type function. The intensity of volume excitation was determined by the *IMFP* database of NIST [I28], and the intensity of surface excitation was a fitting parameter. Coefficients of Drude functions were fitting parameters, too. During the simulation of meaured spectra these fitting parameters had been changing, until the experimental and the simulated data agreed well. For a given sample (e.g. *polySi*), both the surface and the voulme energy loss functions were fixed int he simulations, independently on the measurement geometries and on the electron energies. The only further fitting parameter was the intensity of surface excitation. Using this method, energy loss functions of *polySi*, *amorphous-Ge* and *microcrystalline Sn* were determined [S1, E1-E2].

Theses

1st point

I developed a Monte Carlo model that is suitable for describing electron transport in a multi-component homogeneous and inhomogenous material. I prepared a software according to the model above.

2nd point

I simulated measured *REELS* spectra of *polySi*, amorphous *Ge* and microcrystalline *Sn* samples with good agreement using the *Monte Carlo* program, considering surface losses. The simulation provided surface and bulk energy loss functions of *Si*, *Ge* and *Sn* samples in the 0-50 eV energy range.

3rd point

I determined the surface excitation parameter (*SEP*) of *Si* and *Ge* samples and compared those to the data of Gergely and Werner. The values obtained by the three different models agreed well.

Publications

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SEP parameter can be calculated as the ratio of the number of elastically backscattered detected eletrons (with primary energy) and the number of detected electrons [I29]. Applying this model I calculated *SEP* values. Each surface and volume excitation was calculated seperately. Elasticallay backscattered electrons were listed in the simulation, thus the *SEP* values were calculated easily for *polySi* and *amorphous-Ge* samples. *SEP* values of *Si* and *Ge* are known from Literature. I compared the *SEP* values determined by using the *Monte Carlo* model to *SEP* data of Gergely [I30] and Werner [I31] in the energy range of 0.2-5.0 keV. *SEP* data agreed well.

I determined *IMFP* data of SiO_2 and Si_3N_4 using *EPES* method. Elastic peak intensities were measured on SiO_2, Si_3N_4 and *Si* samples using *Si* reference sample. The *IMFP* data of *Si* were taken from the NIST *IMFP* database [I28]. I determined *IMFP* values from the ratios of SiO_2 / *Si* and Si_3N_4 / *Si* elastic peak intensities. These *IMFP* data are called non-corrected *IMFPs*. I made correction on measured elastic peak intensity data of *Si* and SiO_2 with the surface correction coefficient using Chen’s formula and materials parameter and Kwei’s material parameter. Thus the corrected *IMFP* data of SiO_2 and Si_3N_4 had been obtained. I compared these corrected *IMFP* data to *IMFPs* calculated from optical data and *TPP-2M* formulae [I29]. I concluded that Chen’s formula and materials pameter can be applied for the evaluaton of these measurements. Using this method, *IMFP* data of SiO_2 and Si_3N_4 for given measurement conditions were dermined [S3].

Besides generally used surface sensitive analytical tools (*AES*,*XPS*), *EPES* depth profiling method can provide detailed information about the sample in special cases [I30]. Such situation may occur when multilayer structure of a binary system is investigated. In *EPES* depth profiling technique the change of the concentration along depth can be measured with the change of elastic peak intensity. My Monte Carlo software can calculate elastic peak intensity, and I made simulations on a binary *Ge-Si* samples. I dealt with two types of samples. The first one, which is called 'ideal sample' is consisted of a periodical *Ge* and *Si* layers with a thickness of 20 *A*. The concentration distribution is given by a step-function. Considering the second type of the samples, which is called 'inhomogeneous sample', I assumed that the concentration of the initially ideal sample had been changed because of ion sputtering. The effect of ion sputtering of different energies the distribution of concentration was calculated by *TRIM* simulation [I31]. For both ideal and inhomogeneous samples the concentration values for layers with given width (2 *A*) were fixed. Concentration data of samples were input data in the simulation. Electron energy and measurements geometry were input data, too. I calculated the elastic peak intensity values summarising the elastic peak intensities of the detected elastically backscattered electrons after one elastic collision and after arbitrary number of elastic collisions. The first one is called 'single scattering' and the second one is called 'multiple scattering' calculation.

I simulated *EPES* depth profiling on ideal sample assuming different electron energies and measurement geometries. I compared single and multiple scattering calculations. Differences between single and multiple scattering changed with changing measurement geometry and electron energy.

I examined the sensitivity of the simulation on physical parameters (energy and geometry) in inhomogeneous samples. I determined optimum electron energy for different measurement geometries. I also examined the sensitivity of depth profiles in the function of electron energy on different measurement geometries (electron spectrometers).