Determination of Inelastic Mean Free Path of High Energy Electrons from Shape Analysis of K-Auger and K-conversion Spectra Emitted from Thin Films

L. Kovér*, S. Tougaard¹, J. Tóth, D. Varga, O. Dragoun², A. Kovalík², and M. Ryšavy³

¹Institute of Nuclear Research of the Hungarian Academy of Sciences
P.O. Box 51, H-4001 Debrecen, Hungary
²Physics Dept., University of Southern Denmark, DK-5220 Odense M, Denmark
³Nuclear Physics Institute, Acad. Sci. Czech Republic, CZ-25068 Řež near Prague, Czech Republic

*kovér@atomki.hu

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Inelastic mean free paths of electrons were determined from QUASES spectral shape analysis of high energy photoinduced K-Auger spectra of Cu, Ni and Co thin films, as well as of Fe K-conversion spectra attenuated by Au overlayers of various thicknesses. The results found from analysis of spectra from different layer thicknesses are consistent and they are compared with available estimations in the literature.

1. Introduction

Quantitative applications of non-destructive electron spectroscopic methods in the high (5-10 keV) energy region are gaining an increasing importance recently, especially in the fields of determining thickness of metallic overlayers in several tens of nanometre region and of analyzing deeply buried interface layers. The availability of inelastic mean free path (IMFP) values — important physical parameters for quantification — for electrons in this energy region, however, is very limited.

A major reason for the lack of experimental data is that the necessary experimental conditions fall far beyond that of conventional surface analytical applications of electron spectrometers. Here we report on a novel method for deriving IMFPs from shape analysis of high energy electron spectra. The method is based on the fact that the spectral shape of the part attributable to inelastically scattered electrons, strongly depends on the value of the IMFP. For shape analysis of high energy electron spectra the QUASES-Tougaard [1,2] model and software package, developed for quantitative surface analysis of 3D nanostructures is used, with the Universal Cross Section [2] for inelastic scattering of electrons in the sample.

2. Experimental

Thin layer XPS and XAES

Cu, Ni and Co thin layers of 5-40 nm thickness were vacuum deposited onto silicon wafers using a d.c. magnetron.

High energy Co, Cu, Ni KLL, Co KLM Auger and Cu Lα, Lβ and Si Lx photoelectron spectra were excited from the thin-film samples using Cu characteristic (Kα1, Kα2) and bremsstrahlung X-rays. The electron spectra were measured by the home built ESA-31 electron spectrometer based on a hemispherical analyzer [3] with ~2.4 eV energy resolution (at 7 keV). Fig. 1 shows the measured Co KLL Auger spectrum.

Deposition rates and film thicknesses were monitored by a quartz crystal microbalance (QCM). Independent experimental values for film thicknesses (Cu, Ni) were obtained using cross sectional transmission electron microscopy (XTEM) and scanning probe microscopy (SPM) [4].

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These Al backings with Au layers were irradiated in a nuclear reactor and the intensity of the 412 keV γ line obtained from the samples and standards was measured using a HP Ge detector. Nuclear Activation Analysis yielded a number of Au atoms evaporated onto a known area of the reference backing from which (assuming bulk density) the average thickness of the absorber layers was determined (see Table 2).

The K-shell internal conversion electrons (with the initial energy of 7.3 keV) of the 14 keV transition in $^{57}$Fe were measured in JINR Dubna using the ESA 50 spectrometer based on retarding sphere with a cylindrical mirror analyzer set to an energy resolution of 7 eV [6].

3. QUASES [1] spectral shape analysis

The applied method (and algorithm) is based on the fact, that due to the inelastic scattering of electrons in the solid, the intensity and shape of the peaks in the electron spectra depend strongly on the depth of origin of the excited electrons. The excitation function $F(E,Ω)$ is [2]:

$$F(E,Ω) = \frac{1}{P_r} \left[ \frac{J(E,Ω)}{P_r} - \int dE J(E,Ω) \right] dx \exp \left( \frac{-2m(E-E)}{P_r} \right) \left( \frac{1}{P_r} \right)$$

where

$$P_r = \int dx f(x) \exp \left( \frac{-x}{\lambda_r \cos θ} \right) \sum(s)$$

and

$$\sum(s) = \frac{1}{λ} \int K(T) e^{-αT} dT$$

with

$$λ(E)K(E,T) = \frac{2T}{(C+T)^2}$$

where $J(E,Ω)$ is the measured spectra; $K(T)$ the "universal" cross section for inelastic scattering [8]; $f(x)$ the depth concentration profile ($x$: distance from the surface); $E$ the electron energy; $Ω$ the solid angle of detection; $θ$: the electron emission angle related to the surface normal; $λ_r$: the inelastic mean free path; $T$ the energy loss; and $B,C$ the constants are.

For homogeneous solids $f(x)$ is a constant and [7]:

$$F(E) = J(E) - \frac{1}{α} \int K(E-E')J(E')dE'$$

$F(E,Ω)$ can be determined from the measured spectrum of a pure elemental sample with
infinite "overlayer thickness" (reference spectrum) and in the case of known \( f(x) \)
(overlayer structure/thickness) \( \lambda_i \) is varied until analysis yields similar intensity and peak
shape to the reference spectrum [4].

4. Results and discussion

The IMFPs determined from the high energy \( K \)-Auger and \( K \)-conversion spectra of
Cu, Ni, Co and Au, using the spectral shape analysis, compared to data available in the
literature, can be found in Tables 1 and 2. Our IMFP data show a good agreement for
different overlayer thicknesses and transitions, except for the thickest layer, where the
deviation is appreciable which is expected. The literature data predict systematically
higher IMFP values, however, the differences are only 10-20 % which is comparable to the
expected error due to the spectrum evaluation and the systematic error of the models used for
estimation of the literature data.

<table>
<thead>
<tr>
<th>Transition (energy)</th>
<th>Sample (thickness, nm)</th>
<th>QUASES</th>
<th>Ref. [8]</th>
<th>Ref. [9]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni ( KLL ) (-6500 eV)</td>
<td>Ni 10 a.r. (10.5)</td>
<td>5.8*</td>
<td>6.3</td>
<td>7.8</td>
</tr>
<tr>
<td>Ni 10 (10.5)</td>
<td>5.9</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ni 20 (20.6)</td>
<td>5.9*</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ni 40 (40.8)</td>
<td>(7.3)* high uncertainty</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cu ( KLL ) (-7000 eV)</td>
<td>Cu 20 (21.2)</td>
<td>5.9*</td>
<td>7.8</td>
<td>7.9</td>
</tr>
<tr>
<td>Co ( KLL ) (6050 eV)</td>
<td>Co 10 (7.7)</td>
<td>5.5</td>
<td>6.5</td>
<td></td>
</tr>
<tr>
<td>Co 20 (15.6)</td>
<td>3.9</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Co 40 (33.6)</td>
<td>4.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Co ( KLM ) (6850 eV)</td>
<td>Co 5 (4.5)</td>
<td>5.9</td>
<td>7.2</td>
<td></td>
</tr>
<tr>
<td>Co 10 (7.7)</td>
<td>5.9</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Co 20 (15.6)</td>
<td>5.9</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Co 40 (33.6)</td>
<td>6.3</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Si(Co) ( Is ) (-6200 eV)</td>
<td>Co 5 (4.5)</td>
<td>5.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Co 10 (7.7)</td>
<td>5.3 (5.6)*</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* from the joint (Co \( KLL \), Si \( Is \)) analysis

Ref. [4]
a.r.: "as received"
Table 2
IMFP values (nm) obtained from the shape analysis of the $^{57}$Fe K-conversion spectra of samples with different thickness Au absorbers

<table>
<thead>
<tr>
<th>Transition (energy)</th>
<th>Sample (Au layer thickness, nm)</th>
<th>QUASES</th>
<th>Ref. [8]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe K-conversion (7350 eV)</td>
<td>Au1 (2.7)</td>
<td>6.3*</td>
<td>5.9</td>
</tr>
<tr>
<td></td>
<td>Au2 (6.4)</td>
<td>5.5</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Au3 (8.2)</td>
<td>5.3</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Au4 (12.6)</td>
<td>5.3</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Au5 (16.5)</td>
<td>5.2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Au6 (20.8)</td>
<td>5.3</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td>5.5</td>
<td></td>
</tr>
</tbody>
</table>

* Using the layer thickness equivalent to 9.5 nm of Co determined from the analysis of the shape of the spectrum from the sample not covered by Au.

* The electrons traveled both in Au and Co and the IMFP for Co is larger.

5. Summary
IMFP values in Ni, Cu, Co and Au for electrons with kinetic energies in the 6-7.5 keV energy range were determined using the QUASES shape analysis of high energy electron spectra induced by X-rays or nuclear decay from thin films of several or several tens nm layer thickness.

The analysis provided consistent results for different film thicknesses and for (in the case of Co) various energy transitions. A reasonable agreement was found with IMFP values estimated previously.

Combining high resolution photoelectron or internal conversion spectroscopy in the energy range above 2 keV with spectral shape analysis proved to be a promising method for determining IMFPs of high energy electrons.

6. Acknowledgements
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7. References

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