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

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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 *2s*, *2p* and Si *1s* photoelectron spectra were excited from the thin-film samples using Cu characteristic ($K\alpha_1$, $K\alpha_2$) 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].

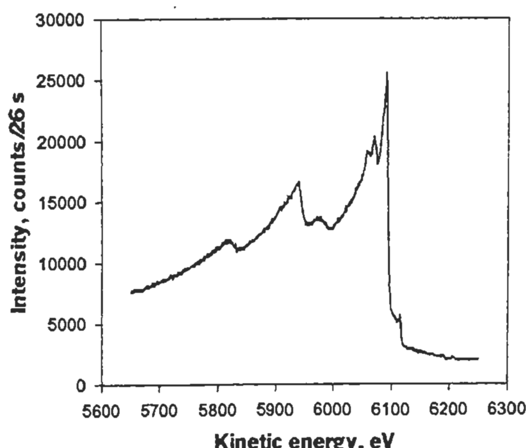

Fig.1 The measured Co *KLL* Auger spectrum excited by Cu X-rays from a 20 nm thick Co layer vacuum deposited onto a Si substrate

Au overlayers [5]

^{57}Co sources were prepared using a standard procedure for purification and electrolytical deposition from solution onto an Al foil of 5 μm thickness. The activity of the resulting ^{57}Co source, determined by gamma-ray spectroscopy, was 2.0±0.1 MBq (~7×10^{13} atoms of ^{57}Co distributed over 50 mm^2). Au layers were successively vacuum deposited onto the ^{57}Co source, inserting always one more clean Al backing into the evaporation apparatus.

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 $F(E,\Omega)$ is [2]:

$$F(E,\Omega)=\frac{1}{P_1}\left\{J(E,\Omega)-\int dE' J(E,\Omega)\int ds\,\exp[i2\pi s(E'-E)]\left(1-\frac{P_1}{P(s)}\right)\right\}$$ where

$$P(s)=\int dx\, f(x)\exp\left[-\frac{x}{\cos\theta}\sum(s)\right];\quad P_1=\int dx\, f(x)\exp\left(-\frac{x}{\lambda_i\cos\theta}\right)$$ and

$$\sum(s)=\frac{1}{\lambda_i}-\int_0^\infty K(T)\,e^{-isT}dT \quad \text{with} \quad \lambda(E)K(E,T)=\frac{BT}{(C+T^2)^2},$$

where $J(E,\Omega)$ 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_i 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]:

$$F(E)=J(E)-\lambda_i\int_E^\infty K(E'-E)J(E')dE'$$

$F(E,\Omega)$ 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) λ_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 1
IMFP values (nm) obtained from the shape analysis of high energy spectra of Cu, Ni and Co thin film samples

Transition (energy)	Sample (thickness, nm)	QUASES	Ref. [8]	Ref. [9]
Ni *KLL*	Ni 10 a.r. (10.5)	5.8$^+$	6.3	7.8
(~6500 eV)	Ni 10 (10.5)	5.9$^+$		
	Ni 20 (20.6)	5.9$^+$		
	Ni 40 (40.8)	(7.3)$^+$	high uncertainty	
Cu *KLL*	Cu 20 (21.2)	5.9$^+$	7.8	7.9
(~7000 eV)				
Co *KLL*	Co 10 (7.7)	5.5	6.5	
(6050 eV)	Co 20 (15.6)	3.9		
	Co 40 (33.6)	4.1		
Co *KLM*	Co 5 (4.5)	5.9	7.2	
(6850 eV)	Co 10 (7.7)	5.9		
	Co 20 (15.6)	5.9		
	Co 40 (33.6)	6.3		
Si(Co) *1s*	Co 5 (4.5)	5.3		
(~6200 eV)	Co 10 (7.7)	5.3 (5.6)*		

* from the joint (Co *KLL*, Si *1s*) 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^{+}

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

$^{+}$ 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.

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