Metal interface formation studied by high-energy reflection energy loss spectroscopy and electron Rutherford backscattering

M. Vos *, M.R. Went

Atomic and Molecular Physics Laboratories, Research School of Physical Sciences and Engineering, The Australian National University, Canberra 0200, Australia

Abstract

We demonstrate that high-energy, high-resolution reflection electron energy loss spectroscopy can provide unique insights into interface formation, especially for the case where an extended interface is formed. By changing the geometry and/or electron energy the electronic structure can be probed over a range of thicknesses (from 10s of Å to more than 1000 Å). At the same time one resolves the elastically scattered electrons into different components, corresponding to scattering of atoms with different mass (so-called 'electron Rutherford backscattering'). Thus these high-energy REELS/elastic scattering experiments obtain information on both the electronic structure and the atomic composition of the overlayer formed.
© 2007 Elsevier B.V. All rights reserved.

1. Introduction

Photoemission is one of the main tools used to study surface layers. Composition can be obtained from core level intensities and the electronic structure can be studied by measuring valence band spectra, or by studying details of the line shape/satellite structures of the core levels. With the advent of synchrotron radiation it became possible to vary the probing depth by tuning the outgoing energies. If surface sensitivity is required one chooses the outgoing energies close to the minimum of the inelastic mean free path. Thicker layers can be studied by increasing the energy of the outgoing electrons. Nowadays, spectra with kinetic energy in the range of 5–10 keV are routinely obtained at the larger storage ring facilities [1]. Decreasing X-ray flux with increasing energy in combination with the decreasing photoexcitation cross section with energy makes any further increase of the probing depth by increasing the photoelectron energy a real challenge.

Reflection energy loss spectroscopy (REELS) has long been used to study the interaction of electrons with a surface, and is an important tool to help understand photoemission in general, as the latter necessarily involves the transmission of electrons from the material under investigation into the vacuum. REELS provides information about bulk and surface loss processes. Indeed by comparing REELS spectra taken at different energies one can determine both the surface and bulk loss functions (see e.g. [2]). At very high energies the surface loss contribution to the spectra becomes rather minor. In this paper we present REELS spectra taken at energies up to 40 keV. As the mean free path of electrons increases with energy this allows us to probe rather thick layers. Here we want to demonstrate that high-energy REELS can be used as an in-house technique to study the electronic structure of thick layers. It is in this sense an alternative to high-energy photoemission, and is a method that could quite easily be adapted to even higher energies.

By comparing the loss functions obtained by high-energy REELS with bulk loss functions, as known from transmission EELS measurements and optical measurements, we

---

* Corresponding author.
  E-mail address: maarten.vos@anu.edu.au (M. Vos).

want to show experimentally that this technique is only affected in minor ways by surface effects. Subsequently we will show examples of interface formation by deposition of an Al film on several substrates (Mo, Pt and Au). Aluminum is chosen as it has a simple energy loss spectrum dominated by a plasmon peak. The energy loss spectra of Mo, Pt and Au are more complicated, but all have characteristic features, easily resolved with our spectrometer. Can the spectra obtained from an overlayer system be described, at least in first-approximation, as a linear combination of the substrate spectra and the overlayer spectra? We will see that this sometimes is the case, but often not. Often Al reacts with the substrate and an extended interface is formed. By adding Al in subsequent evaporations we can establish at what point pure Al forms, as is evident of the appearance of the Al plasmon. This signals the end of the interface formation. It can happen for surprisingly large Al layer thicknesses (several 100s of Å), and the bulk sensitivity of this technique is thus very beneficial. High-energy REELS is thus shown to be a good technique to obtain information on thick layers (at least by conventional electron spectroscopy standards).

Interest in investigating the possibilities of these high-energy REELS measurement was triggered in part by the realization that the elastic peak splits up at high energies in different components, corresponding to electrons scattering of atoms of different mass [3]. This is because if an electron is scattered over a large angle, it transfers a significant amount of momentum to the scattering atom. If the momentum transferred to the atom is **q**, then the corresponding (mean) recoil energy E_r, transferred from the electron to the atom is given by

$$E_r = q^2/2M \tag{1}$$

with M the mass of the atom. The magnitude of E_r for the element studied in the paper is given in Table 1 together with other scattering properties. The electron energy will be reduced by this amount. Thus, in favorable cases, the elastic peak splits up into different components, due to scatterers with different mass M and the measured structure can be used to determine the surface composition. This technique is then often referred to as 'electron Rutherford backscattering' (ERBS) (see e.g. [4]), as it resembles (ion) Rutherford backscattering in many ways. REELS at high energies provides complimentary information, mainly on the electronic structure, that is obtained simultaneously with ERBS data. In spite of the different acronyms the ERBS experiment is identical to a REELS experiment taken at high energies. In REELS one obtains information of the part of the spectra that is due to electronic excitations *in addition* to elastic scattering, in ERBS one considers the (quasi-)elastic peak only. Sometimes, as we will see, one has also to consider the influence of the recoil energy on the shape of the energy loss structures [5].

The first attempts to use ERBS as an analytical tool were made, not surprisingly, for the case of hydrogen, as, due to its small mass, the recoil energy for protons is rela-

Table 1
Various calculated quantities pertaining to elastic scattering for different elements at different incoming energy E_0

Element	E_0 (keV)	E_r 120° (eV)	$\frac{d\sigma}{d\Omega}$ 120° (Å^2)	λ_{el} (Å)	λ_{tr} (Å)	λ_{in} (Å)
Al	5	0.30	1.70×10^{-4}	40	4950	88
Mo	5	0.09	2.12×10^{-3}	15	319	59
Pt	5	0.04	3.02×10^{-3}	13	147	41
Al	20	1.24	1.00×10^{-5}	170	21,900	288
Mo	20	0.35	1.37×10^{-4}	39	2590	190
Pt	20	0.17	5.95×10^{-4}	28	980	131
Al	40	2.53	2.44×10^{-6}	307	74,000	528
Mo	40	0.71	3.32×10^{-5}	64	7950	346
Pt	40	0.35	1.70×10^{-4}	42.2	2800	237

The recoil energy E_r was obtained from Eq. (1). The differential elastic scattering cross section $\frac{d\sigma}{d\Omega}$ at 120° and 40 keV was obtained from the ELSEPA package [6] and so was the elastic mean free path $\lambda_{el} = 1/N\sigma_{el}$ (with N the atomic density and σ_{el} the total elastic cross section) and the transport mean free path λ_{tr}. The inelastic mean free path λ_{in} was obtained from the TPP formula [7].

tively large. At high energies, using Rutherford cross sections, there were systematic deviations from the expected and observed H peak area [8]. At lower energies, with the H peak only partly resolved, Yubero et al. used empirical cross sections obtained from the measurements of various plastics to determine the H concentration in amorphous carbon films, and they obtained reasonable agreement with ion beam analysis results [9]. By increasing the energy and scattering angle it became possible to separate many more elements. For example we could separate Si from O, S from Mo, etc. [4]. On the other hand, when studying samples containing e.g. Fe and Au, then the Fe peak is only partly resolved. Even when studying well-defined compounds the observed elastic peak intensity often deviates by 20% from the expected values [4]. Thus interpretation of ERBS data on a fully quantitative level is still in its infancy. Interpretation for reactive overlayer systems is even more complicated than for stoichiometric compounds [10], as in this case the thickness of the overlayer and reacted layer, as well as their inelastic mean free path are required.

There is a strong dependence on the elastic cross section on the atomic number (it scales roughly with Z^2). For overlayer systems this means that the probing depth can vary greatly, depending on the elemental depth distribution [11–13]. For some of the cases studied here the probing depth is much larger than the inelastic mean free path. In this paper we will restrict ourselves to semi-quantitative interpretation of the elastic part of the spectrum, mainly to discuss the probing depth in these experiments.

High-energy EELS spectra are usually taken in a transmission geometry. This technique was developed into a fine art the 1960s and remarkable energy resolutions were obtained (e.g. 50–80 meV at 50 keV [14]). It then became integrated in the electron microscope, and is nowadays one of the main techniques in quantitative electron microscopy [15]. EELS in a reflection geometry is usually done at $\simeq 1$ keV energy with modest resolution ($\simeq 1$ eV). REELS Measurements up to 10 keV were reported by Tougaard