Available online at www.sciencedirect.com
ScienceDirect
Surface Science 601 (2007) 2270–2276
SURFACE SCIENCE
www.elsevier.com/locate/susc

Joint experimental and computational study of aluminum electron energy loss spectra

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Received 21 December 2006; accepted for publication 21 March 2007
Available online 27 March 2007

Abstract

Experimental reflection electron energy loss (REEL) spectra are measured from aluminum for primary energies ranging from 130 eV to 2 keV. A Monte Carlo simulation is shortly described and used to calculate the same spectra. The focus is on reproducing the variable weight of surface and bulk losses as the surface sensitivity of spectra changes by changing the primary electron energy. The intensity of surface losses in the simulations is modulated by the thickness of the region where surface excitations occur. Simulations based either on a constant or an energy-dependent thickness for this layer are considered. In both cases, simulated spectra reproduce the experimental trend as a function of energy, though the correct surface-to-bulk intensity ratio for each energy is either underestimated or overestimated.

Keywords: Aluminum; Reflection electron energy loss spectroscopy; Monte Carlo simulation; Impact phenomena; Plasmons; Surface excitations

1. Introduction

While the interaction of electrons with energy higher than 10 keV with solid targets is well known, the low energy processes, elastic and inelastic, need further investigations. The Monte Carlo strategy is today the best approach in the calculation and prediction of radiation effects in solids. It requires an accurate description of the elastic and inelastic scattering processes which occur during the travel of the particle inside the solid target.

A quantitative analysis of the elastic scattering cross-section, for low energy electrons in solids ($E_0 < 10$ keV), is badly approximated in the first Born approximation, and requires instead an accurate numerical solution of the quantum-relativistic equations governing the electron interactions with a screened central potential (Dirac–Hartree–Fock–Slater atomic potential). Furthermore, exchange and solid state effects should be introduced in the calculations.

Regarding the inelastic scattering properties, the use of the Bethe stopping power formula is not adequate for low energies and the dielectric approach is mandatory. Furthermore, the use of a stopping power is, generally, questionable, even if derived in the dielectric function scheme, because it corresponds to the continuous slowing down approximation. While such an approximation can be safely used for the calculation of the absorption and of the backscattering coefficients, it cannot be utilized for the simulation of energy loss spectra because it neglects the fact that the energy is actually lost in discrete collisions, that the number of such collisions statistically fluctuates and that the electrons, along their tracks, occasionally suffer very large energy losses in close collisions with the atomic electrons. On the other hand, the statistical fluctuations of energy losses are adequately taken into account by using the energy straggling strategy.

The aim of this work is to investigate the elastic and inelastic scattering properties of electrons traveling in the surface layers of aluminum.

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doi:10.1016/j.susc.2007.03.029

Aluminum properties have been extensively studied (see, for example, [1] and references therein) and the interaction of electron beams with Al has been theoretically and numerically investigated [2–4]: nevertheless several questions are still open.

Electron beams are widely used in analytical techniques for the chemical characterization of materials and – for quantitative applications of the electron spectroscopies – a theoretical understanding of the microscopic processes occurring while electrons travel in the solid target plays a crucial role.

A number of phenomena can be experimentally observed by REEL spectroscopy. In this paper, we present a comparison between our experimental and computational results concerning, in particular, the behavior of surface and bulk-plasmon peaks in Al as the energy of incident electrons changes from 130 to 2000 eV.

2. Experimental

Experimental REEL spectra were measured, in the reflection mode, within a PHI545 instrument equipped with a double-pass cylindrical mirror analyzer and a coaxial electron gun. Primary beam energies ranging from 130 to 2000 eV were considered. The angle between the sample surface normal and the incident electron beam was 30°. All REEL spectra were taken at a constant energy resolution of 0.6 eV, as measured on the Pd Fermi edge of a HeI ($h\nu = 21.2$ eV) excited valence band photoemission spectrum. An aluminum foil was sputtered in Ultra High Vacuum with a 4 keV Ar^+ ion beam until no oxygen was detected by AES (Auger electron spectroscopy). Once acquired, the REEL spectra were corrected for the energy dependence ($E^{-0.9}$) of the analyzer transmission function.

3. Theory

3.1. Inelastic scattering

From the Drude theory, designed to model the interaction of optical radiation with a dilute gas of free electrons, it follows that the frequency dependent dielectric function ε, i.e., the ratio between the electric displacement $\mathscr{D}$ and the electric field $\mathscr{E}$, is given by [5]

$$\varepsilon(\omega) = \frac{\mathscr{D}}{\mathscr{E}} = 1 - \frac{\omega_p^2}{\omega^2 + i\gamma\omega}, \tag{1}$$

where γ is a damping constant, and ω_p is the plasma frequency

$$\omega_p = \sqrt{\frac{nNe^2}{\varepsilon_0 m}}. \tag{2}$$

Here n is the number of valence electrons per atom, N is the number of atoms per unit volume, e and m are the electron charge and mass, and ε_0 is the vacuum dielectric constant.

Within the framework of the dielectric theory, the spectral response of the solid in inelastic scattering experiments is described by the so called *energy loss function* which, in the optical limit (i.e., for zero momentum transfer), is given by

$$f(\omega) = \mathrm{Im}\left[-\frac{1}{\varepsilon(\omega)}\right]. \tag{3}$$

[1]Since aluminum can be considered, to a good approximation, a free electron metal, we use Eq. (1) to describe its response to the electric field associated with travelling electrons. In particular, by allowing for different damping constants in the bulk and in the surface layer, we introduce a bulk optical dielectric function

$$\varepsilon_B(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\gamma_B\omega}, \tag{6}$$

and a surface optical dielectric function

$$\varepsilon_S(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\gamma_S\omega}. \tag{7}$$

The plasma energy, $\hbar\omega_p = 15.3$ eV ($\hbar = h/2\pi$, with $h =$ Planck constant), and the damping constants, $\hbar\gamma_B = 1.8$ eV and $\hbar\gamma_S = 2.7$ eV, have been determined in order to obtain the best agreement with our 2 keV experimental data. It is worth noting that $\hbar\omega_p = 15.3$ eV is lower than $\hbar\omega_p = 15.8$ eV which would be derived, for the Al mass density, from Eq. (2). The lower plasmon energy reflects a lower Al mass density for our sample as compared to bulk, crystalline Al. Since the sample was sputter-cleaned, we ascribe the lower mass density to sputter-induced amorphization in the surface region.

To properly describe electron inelastic scattering, relations (6) and (7) should be extended from the optical limit to non-zero momentum transfer $\mathbf{k}$ by introducing dielectric functions $\varepsilon_B(\mathbf{k},\omega)$ and $\varepsilon_S(\mathbf{k},\omega)$ which depend on both energy and momentum transfer. The bulk and surface response of the solid is then described by the bulk and surface electron energy loss functions

$$f_B = \mathrm{Im}\left[-\frac{1}{\varepsilon_B(\mathbf{k},\omega)}\right], \tag{8}$$

$$f_S = \mathrm{Im}\left[-\frac{1}{\varepsilon_S(\mathbf{k},\omega)+1}\right]. \tag{9}$$

The $\mathbf{k}$ dependence of ε_B and ε_S can be factorized by introducing the Ashley function [7]

[1] Note that, in the notation of Yubero and Tougaard [6], the dielectric function is given by

$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2 - i\gamma\omega}, \tag{4}$$

and hence,

$$f(\omega) = \mathrm{Im}\left[\frac{1}{\varepsilon(\omega)}\right]. \tag{5}$$