Available online at www.sciencedirect.com
ScienceDirect
Journal of Electron Spectroscopy and Related Phenomena 159 (2007) 62–65
JOURNAL OF ELECTRON SPECTROSCOPY and Related Phenomena
www.elsevier.com/locate/elspec

Effective energy loss functions of Mo and Ta derived from reflection electron energy loss spectra

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Received 13 January 2007; received in revised form 6 March 2007; accepted 10 March 2007
Available online 15 March 2007

Abstract

Effective energy loss functions (EELF) of Mo and Ta were derived from reflection electron energy loss spectroscopy (REELS) spectra measured at different primary energies using extended Landau approach. The plasmon behaviors and fine structures of energy losses presented in the energy loss function are analyzed. For EELF of Mo, the peak at 9.9 eV is a composition of bulk plasmon at 10.4 eV and surface plasmon at 9.5 eV, and the other peak at 22.4 eV is made of bulk plasmon at 24.4 eV and surface plasmon at 19.8 eV. As for Ta, the peak at 19.7 eV is contributed by bulk plasmon at 20.8 eV and surface plasmon at 8.4 eV. The EELF thus includes both the surface effects and bulk excitation, where surface excitation is important mainly at low energy losses; the competition between surface and bulk excitation modes is analyzed for varied primary energy. The Monte Carlo simulated REELS spectra with the derived EELF agree well with the experimental results.

PACS: 34.50.Bw; 34.80.Dp; 71.45.Gm; 79.20.Uv

Keywords: Reflection electron energy loss spectroscopy; Effective energy loss function; Molybdenum; Tantalum

1. Introduction

Energy loss processes of signal electrons in surface sensitive electron spectroscopies, such as X-ray photoelectron spectroscopy (XPS) and reflection electron energy loss spectroscopy (REELS), have been attracting much attention. When a primary electron impinges and penetrates into a solid or a signal electron escapes, it suffers certain energy loss through inelastic scattering process both inside the solid and in the surface/vacuum region before either being detected as signals or coming to rest inside the solid by losing all of its kinetic energy.

Theoretical treatments to the inelastic scattering process and the energy loss spectra of emitted electrons were based on dielectric response theories [1–3]. Yubero et al. have developed a simple model to calculate the effective cross-sections in order to determine inelastic scattering sections from REELS spectra [4,5]. Many authors have pointed out that, without considering surface effect a large discrepancy would exist when comparing theoretical and experimental spectra of low losses and particularly for low primary energies [6–8]. Surface excitation effect is important for quantitative surface analysis and for experimental evaluation of electron inelastic mean free paths (IMFPs) from elastic peak spectroscopy.

A variety of theoretical and experimental studies on the surface excitation phenomenon have been reported [9,10,13,14], with different levels of complexity for evaluation and quantification of surface effects. Chen et al. have taken into account additional inelastic scattering probability due to surface effect by use of a surface excitation parameter (SEP) [9]. Ding and co-worker have also investigated the surface effect in surface electron spectroscopy with a formalism of electron self-energy [10–12]. Werner has extracted SEP from experimental REELS spectra of several metals and semiconductors at medium primary energies by decomposing REELS spectra into contributions

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doi:10.1016/j.elspec.2007.03.006

from surface and bulk excitations; surface excitation component was obtained by means of a combined elimination-retrieved algorithm, and, exact reversion of a bivariate power series in his treatments [14,15]. Although these methods describe experimental results quite well, they are generally very complex and expatiatory in form and, thus, are hardly applicable to practical analysis.

On the other hand, Yoshikawa et al. obtained an effective energy loss function (EELF) which contains both the surface and bulk excitations from the measured REELS spectra by an extended Landau approach [7,16]. The EELF including surface and bulk excitations is convenient for use in practice. Furthermore, instead of iterative deconvolution method that used by Yoshikawa et al. Zhang et al. obtained a formula for numerical multi-fold self-deconvolution so that the EELF can be more easily evaluated from a measured REELS spectra [8].

In this paper, the EELFs of Mo and Ta are obtained by this extended Landau approach. They are then compared with theoretical surface and bulk energy loss functions derived from the optical constant. In order to verify the derived EELF the REELS spectra are reproduced by a Monte Carlo simulation with EELF. Finally, the competition between surface and bulk excitation modes as presented in EELF is analyzed for varied primary energy.

2. Theoretical

The EELF can be derived from a measured REELS spectrum using the extended Landau method. Landau formulation in the Fourier space of the energy is represented as

$$\tilde{J}(s) = \tilde{F}(s)\sum_{n=0}^{\infty}\alpha_n[\lambda_{\text{in}}\tilde{K}(s)]^n \tag{1}$$

where $\tilde{J}(s)$, $\tilde{F}(s)$, $\tilde{K}(s)$ are the Fourier transforms of the measured REELS spectrum $J(E)$, the energy distribution of primary electrons $F(E)$, and the differential inverse inelastic mean free path (DIIMFP) $K(\Delta E)$, where ΔE is the energy loss, respectively. λ_n is the IMFP, s the Fourier parameter conjugate to the energy, α_n the correction factor for taking into account the elastic scattering effect and n is the number of inelastic scattering events. Correction factor α_n determines how the electrons undergoing inelastic scattering events in a sample contribute to the REELS spectrum. The term $[\lambda_{\text{in}}\tilde{K}(s)]^n$ represents the contribution to the spectrum of the detected electrons suffering n-fold inelastic scattering events before emission from the sample surface.

Using the experimental $\tilde{J}(s)$ and $\tilde{F}(s)$ and the theoretical value of α_n, $\tilde{K}(s)$ or $K(\Delta E)$ is then derived from Eq. (1). The absolute value of the DIIMFP is determined by the following equation:

$$\int_0^{\infty}\lambda_{\text{in}}K(\Delta E)\text{d}(\Delta E) = 1 \tag{2}$$

In the present study, we used the reference IMFP values reported by Tanuma et al. [17]. Finally, the EELF, $\text{Im}\{-1/\varepsilon_{\text{eff}}(\omega)\}$, is numerically obtained by solving the following integration:

$$K(\Delta E) = \frac{1}{2\pi a_0 E}\int_0^{\infty}\frac{\hbar\omega}{\Delta E(\Delta E-\hbar\omega)}\,\text{Im}\left\{\frac{-1}{\varepsilon(\omega)}\right\}\text{d}(\hbar\omega)\Theta \times\left[\frac{\hbar^2}{2m}(2k\bar{q}-\bar{q}^2)-\Delta E\right] \tag{3}$$

where ΔE and $\hbar\omega$ represent the energy loss of an electron while $\hbar\omega$ is an integration variable, a_0 the Bohr radius, E the electron kinetic energy and $\Theta(x)$ is the step function representing the momentum and energy conservation laws. The above equation is derived [18] by extending the ω-dependent optical energy loss function, $\text{Im}\{-1/\varepsilon(\omega)\}$, into (q, ω)-space with Penn's algorithm [2]. $\hbar\bar{q}$ is the momentum transfer satisfying the following dispersion relation:

$$\Delta E = \hbar\omega + \frac{(\hbar\bar{q})^2}{2m} \tag{4}$$

Using the optical dielectric constants $\varepsilon(\omega)$, Eq. (3) yields just the DIIMFP for bulk excitation. The function $\text{Im}\{-1/\varepsilon_{\text{eff}}(\omega)\}$ derived by the present extended Landau approach, however, should include all energy loss processes in which not only bulk excitation but also surface excitation is involved. This is the reason it is called the EELF.

3. Experimental

The REELS spectra were measured with a cylindrical mirror analyzer (CMA) equipped with a coaxial electron gun. In this system, signal electron current was measured with a Faraday cup. The energy resolution of the system is $\Delta E/E = 0.25\%$. The measurement was performed at primary energies of 500, 1000, 1500 and 2000 eV electrons impinging on Mo and Ta samples at normal incidence. The samples surface was cleaned by 250–300 eV Ar^+ sputtering. Details of the measurement system have been described elsewhere [19]. CMA only detects those electrons coming into the solid corn by the angular aperture from 36.3° to 48.3°. Hence, the Monte Carlo simulation was performed to match exactly the experiment by counting only those reflected electrons that are detected by the CMA.

4. Results and discussion

Fig. 1 shows the measured REELS spectra for Ta and Mo at the primary energy of 1 keV. Several peaks appear in the spectra for the two types of transition metals due to the non-localized d electrons. REELS spectrum for monochromatic primary energy is then obtained by a deconvolution with a Gaussian elastic peak shape, using the numerical formula for extended Landau approach as described by Zhang et al. [8,20]. The EELFs extracted from these experimental REELS spectra are shown in Fig. 2 and are compared with the surface energy loss function (SELF), $\text{Im}\{-1/[1+\varepsilon(\omega)]\}$, and the bulk energy loss function (BELF), $\text{Im}\{-1/\varepsilon(\omega)\}$ that obtained from optical dielectric constants [21]. It is easily seen that the EELF includes both surface and bulk excitations. The surface effect dominates at low energy