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Modelling the directional and energy dependence of 5–10 keV Ar^+ ion-induced secondary electron yields from Cu crystals

M.A. Karolewski *, R.G. Cavell

Department of Chemistry, University of Alberta, Edmonton, AB, T6G 2G2, Canada

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ABSTRACT

The primary ion directional effects observed in secondary electron yields induced by ion bombardment [5 keV $Ar^+ \rightarrow Cu(100)$] are simulated using a semi-empirical molecular dynamics model. The directional effects are presumed to arise from inelastic energy transfers that take place in close binary atomic encounters. The latter are estimated using the Oen-Robinson model, in combination with a critical apsidal distance (R_c). The connection between the measured kinetic electron emission (KEE) yields (γ_{KEE}) and the predicted inelastic energy loss in a binary atomic collision (ΔE_i) is established through a semi-empirical fitting procedure involving R_c and other parameters in the following model: $\gamma_e = \gamma_0 + \gamma_{KEE} = \gamma_0 + \langle \Delta E_i(z) \exp(-z/\lambda) \rangle$, where z is the collision depth. The directional effects are best reproduced by fitting the model to Ar–Cu inelastic collisions for two azimuthal incident directions: R_c is estimated to be 0.47 ± 0.03 Å; the parameter, λ (an effective electron attenuation length), is estimated to be 18 ± 2 Å. The same model also describes the γ_{KEE} energy dependence for 5–10 keV Ar^+ normally incident on low-index Cu crystal targets [Phys. Rev. 129 (1963) 2409]. The spatial and temporal distributions of the hard collisions that initiate KEE are discussed on the basis of the model.

1. Introduction

Several distinct mechanisms may contribute to ion-induced electron emission [1,2]. Potential electron emission (PEE) is a consequence of ion neutralisation, while kinetic electron emission (KEE) refers to other emission mechanisms that display an ion energy threshold. The primary excitations that initiate KEE are either non-adiabatic electron promotions that are stimulated by close binary atomic encounters ("promotional" KEE), or kinetic energy transfers to the electron gas in the form of single-particle or collective excitations ("electronic KEE") [3,4]. Various other de-excitation and energy loss processes may contribute to the KEE yield, such as Auger electron emission, plasmon decay, and inelastic electron scattering [5].

KEE yields from single crystals exhibit prominent modulations when the ion direction of incidence is varied [6,7]. The modulations can also be observed in the target current [8], and hence can be exploited for rapid alignment of crystals in ion beam experiments [9,10]. The modulations originate from lattice transparency effects that control the probability of hard collisions between projectile and target atoms. Von Gemmingen measured the critical impact parameter for KEE in the H–Ni system [11]. Soszka [12] and Rabalais et al. [13] drew attention to the directional similarity of KEE and backscattered ion yields. Bernhard et al. [14] deduced the alignment of atomic rows in metal overlayers from the azimuthal variation of the target current.

The simulation of directional effects in KEE yields has a long history [6]. The earliest models were based on hard-sphere scattering, and the inelastic energy transfer was assumed to vary in proportion to the elastic energy transfer [15,16]. Subsequently Martynenko formulated an analytical theory of directional effects that incorporated the Parilis-Kishenevsky (PK) theory [17] for computation of the inelastic energy transfers leading to electronic excitations, and also employed shadowing concepts [18]. Harrison et al. [19] developed a single collision model of KEE yield variations with surface crystallographic orientation that utilised the Firsov inelastic theory [20].

This paper models the directional effects observed in ion-induced (5 keV Ar^+) target currents from Cu(100), by equating the relative number of KEE excitations to the mean inelastic energy loss predicted within a molecular dynamics (MD) framework. The MD simulations are constructed on the presumption that the directional effects arise from inelastic energy transfers that take place in close binary atomic encounters, characterised by a threshold or critical apsidal distance, R_c, which can be determined by fitting a semi-empirical model to experimental data (the apsidal distance refers to the distance of closest approach that is attained at the apsis, or turning point, of a binary collision). It is also shown that the same model can reproduce the previously reported energy dependence of KEE yields for different low-index Cu surfaces and 5–10 keV Ar^+ primary ions.

To date, MD has not been used for simulations of KEE directional effects, probably due to the high computational costs required for

* Corresponding author at: School of Education, University of New South Wales, Sydney, NSW 2052, Australia.
E-mail address: karolewski@alum.mit.edu (M.A. Karolewski).

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angular profiles. However, binary collision approximation (BCA) simulations have been used to model directional effects in ion-induced Auger and secondary electron emission [21–23]. These BCA studies employed semi-empirical models of the primary inelastic excitation processes. In Refs. [21,22], excitation was initiated upon attainment of a critical apsidal distance, while in Ref. [23] the excitation energy was computed as a local inelastic loss based on the Oen-Robinson (OR) inelastic model [24]. More recently, various groups have developed atomistic methods for simulating inelastic energy transfers that are derived from fundamental principles [25–31], and in some cases include inelastic energy transfer mechanisms that are initiated upon attainment of a critical apsidal distance in close binary encounters.

2. Experimental

The experimental target current measurements were performed using an ultrahigh vacuum (UHV) secondary ion mass spectrometer (base pressure 10^{-10} mbar) that was equipped with a rastered, microfocussed VG AG61 filament ion source. Electrostatic deflection of the ion source beam reduced the target current to <0.1% of the primary beam current, indicating an absence of neutral species in the primary beam. The ionisation grid is held at 50 V with respect to the filament. At this energy, the cross-section for production of Ar^{2+} is 0.7% of that for Ar^{+}[32].

The Cu(100) substrate was oriented by Laue diffraction to better than 1°, then mechanically polished (1 μm), chemically etched (HNO_3) and rinsed (water, 2-propanol) prior to insertion in the vacuum system. After bakeout, the cleaning of the crystal in UHV involved numerous cycles of Ar^{+} bombardment and heating (~1000 K), which reduced all contaminants to satisfactory levels, as determined by SIMS (e.g. for 1 nA Ar^{+}: C_2^-, CN^-, Cl^- <50 counts s^{-1}; O^-, C_2H^- <10 counts s^{-1}). For comparison, ~0.5 monolayers coverage of an atomic electronegative species, X, would produce ~3×10^4 counts s^{-1} of the X^- or X_2^- species when X = C or Cl, and ~130 counts s^{-1} when X = O [33].

The ion-induced target current was measured using a Keithley 485 picoammeter interposed between the target and ground. Measurement of the primary current was carried out using a 50 V battery placed in series, to provide a positive bias for suppression of low-energy electrons. The primary ion current used for the measurements was 0.3 nA, while the mean primary current density was ~3 nA cm^{-2}. The target current angular scan was completed in 300 s, corresponding to an ion fluence <10^{13} cm^{-2}. The target current (I_t) is the sum of primary ion current (I_p) and the secondary electron current, so the secondary electron yield coefficient (γ_e) can be estimated from the relationship:

$$I_t = I_p(1 + \gamma_e). \tag{1}$$

Contributions to the measured current from tertiary electrons (liberated from the chamber walls by reflected ions) were not found to be significant for the geometries used in these experiments, although they can be detected (via I_p measurements) at near-grazing incidence. Electron yield coefficients derived from target current measurements have been shown to agree with those from other methods to within experimental errors (typically 5%) [34,35]. A reviewer has pointed out that Eq. (1) is based on the assumption that contact potential differences between the target (nominally at ground) and the surrounding electrodes do not impede efficient transport of low-energy secondary electrons away from the target. We assume that any possible effect of this kind on the derived electron yield is small.

This study follows the angular convention used in low-energy ion scattering, whereby the altitudinal (ψ) angle is used to describe the projectile angle of incidence in the plane lying perpendicular to the surface. The sample manipulator permitted accurate rotation (±0.5°) of the altitudinal angle, and uncalibrated but continuous rotation of the azimuthal (ϕ) angle. The procedure for aligning the crystal with respect to the ion gun was similar to that described in [9]. The target normal direction ($\psi = 90°$) was aligned with the ion beam by exploiting the fact that the electron yield (hence target current) passes through a minimum at normal incidence. The target azimuthal orientation was then aligned with respect to the ion beam by azimuthal rotation at a tilt angle $\psi = 29°$. In this geometry, prominent target current minima are observed for the <001> ($\phi = 0°$) and <021> ($\phi = 26.6°$) azimuthal directions.

3. Computational method

MD simulations of the directional dependence of inelastic energy losses from Ar^{+}–Cu collisions were performed with the *Kalypso* software package (version 3.1) [36]. For the Cu(100) simulations, the Ar^{+} azimuthal (ϕ) angle of incidence was parallel to either <001> or <021>, while the altitudinal (ψ) angle of the incident projectile was varied between 25° and 90° (normal incidence). The Cu(100) target crystallite consisted of 18 Cu(100) atomic layers (each having dimensions 128×83 Å^2; ca. 27,000 atoms in total). The side faces of the crystallite were terminated by free (001) edges; the surface layer was relaxed inwards by 1.7%. Random, uncorrelated, Debye–Waller vibrational displacements were applied to each target atom, assuming a temperature of 300 K, and standard surface and bulk Debye temperatures [37,38]. For each (ϕ, ψ) direction of incidence, 784 Ar^{+} projectile trajectories were simulated. The dimensions of the primary impact zone were 1.807×1.807 Å^2 and 3.615×3.615 Å^2 for <001> and <021> incidence, respectively. Normal Ar^{+} incidence simulations were also performed for Cu(110) and Cu(111) targets, using crystallites of comparable dimensions and appropriate modifications of the primary impact zone shape and dimensions.

The attractive part of the Cu–Cu interaction was described using a many-body tight-binding (TB) potential [39]. A switching function was applied to the potential for r>3.62 Å that terminates the potential and force functions smoothly at the cut-off distance (4.43 Å). The Cu–Cu TB potential was interpolated at short internuclear distances to a repulsive Bohr potential fitted to ab initio data [40]. A Bohr potential fitted to ab initio data [40] was also used for the Ar–Cu interaction at all separations below the same cut-off.

Inelastic energy losses were predicted using the Oen-Robinson (OR) inelastic loss function, in which a discrete inelastic energy loss is computed as a function of apsidal distance (distance of closest approach) for each binary collision [24]. The OR excitation function estimates the inelastic energy loss (ΔE_i) arising from a binary atomic collision, in which the apsidal distance is R_0, as follows:

$$\Delta E_i = \frac{0.09e^2 a_B v}{\pi\varepsilon_0 a^2 v_B} \frac{Z_1^{7/6} Z_2}{\left(Z_1^{2/3} + Z_2^{2/3}\right)^{3/2}} \exp(-0.3R_0/a) \tag{2}$$

where a is the Molière–Lindhard screening length; a_B is the Bohr radius; v_B is the Bohr velocity; e is the proton charge; ε_0 is the vacuum permittivity; Z_1 is the atomic number of the projectile (or recoil) species; Z_2 is the atomic number of the target atom species; v is the asymptotic relative speed of the collision partners, which is calculated as follows:

$$v = \left[v_a^2 + 2V(R_0)/\mu\right]^{1/2} \tag{3}$$

where v_a and $V(R_0)$ represent, respectively, the magnitude of the relative velocity, and the potential energy of the collision pair, at the apsidal point, while μ is the reduced mass of the collision system.

The OR excitation function predicts total electronic stopping cross-sections of similar magnitude to those of the Lindhard–Scharff–Schiøtt (LSS) theory [41], but chooses the spatial dependence of the inelastic loss function to reflect approximately the electron density around the target nuclei. Both models predict that relative electronic