Electron emission from a metal surface bombarded by slow highly charged ions

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We have measured total electron-emission yields γ for clean tungsten bombarded with various multiply charged ions: Z^{q+} up to N⁶⁺, Ne⁷⁺, Ar¹²⁺, and Kr¹¹⁺. At low impact energy (≤ 10 eV/amu) a linear dependence of γ versus ion potential energy could be observed up to the respectively highest q, for which a pronounced decrease appeared superimposed on the gradual decrease of all γ toward higher impact energy. Our results are discussed within a multiple-step ion-neutralization model.

Electron emission from a metal surface under bombardment of multiply charged ions Z^{q+} can be caused by two basically different mechanisms.

(1) Potential emission (PE) results from interaction of empty projectile states with the surface valence-band states. One can specify contributions from Auger neutralization, Auger deexcitation after resonance neutralization (RN-AD), and for multiply charged ions also autoionization after multiple resonance capture (RN-AI).¹⁻³ The RN processes can already proceed at rather large distances from the surface, and in principle no kinetic energy of the projectile is needed for PE to take place.

(2) Kinetic emission (KE) is initiated in close encounters between projectile ions and target particles with the kinetic energy of the projectile dissipated such that slow electrons are emitted from the target.⁴ To a small extent also Auger electrons can be produced from inner shell vacancies.⁵ KE can only proceed beyond a certain threshold impact velocity v of typically 10⁵ m s⁻¹, above which both PE and KE contribute—probably independently—to the apparent electron emission yield

 $\gamma_q = \gamma_{q,\text{PE}} + \gamma_{q,\text{KE}}$.

According to available experience, γ_{PE} decreases with increasing v,^{6,7} and at fixed ion-impact velocity it seems to depend linearly on the total potential energy W_q carried by the ion toward the surface:^{2,8}

$$\gamma_{q,\text{PE}} \approx k W_q$$
, with $W_q \equiv \sum_{i=1}^q I_{i-1,i}$, (1)

where $I_{i-1,i}$ is the ionization potential of ion $Z^{(i-1)+}$.

Relation (1) was shown to hold for impact of multiply charged rare-gas ions on both Mo and W up to $W_q \approx 450$ eV [Xe⁸⁺, $\gamma \approx 7.5$, with $k \approx 1.7 \times 10^{-2}$ eV⁻¹ (Ref. 8)] as

well as for impact of various metal ions on Al and Cu-Be up to $W_q \approx 750$ eV [In⁹⁺ on Cu-Be, $\gamma \approx 9$, with $k = (0.8-2.6) \times 10^{-2}$ eV⁻¹].⁷ If, however, KE dominates (especially for high v and low q), there is no pronounced qdependence of γ .^{9,10}

So far, neutralization of multiply charged ions at a solid surface is neither understood in detail nor has it been broadly studied, most probably because of experimental limitation. It is quite tempting to test the validity of relation (1) toward considerably higher values of W_q than so far involved, since for high q rather strong electron emission is predicted, being possibly accompanied by interesting new phenomena. Recently such testing for $W_q \gg 1000$ eV has become feasible with the availability of powerful novel multiply charged ion sources of the electron-cyclotron-resonance ion-source (ECRIS) type,¹¹ from which well-defined beams of slow, highly charged ions can be produced via simple ion deceleration.

Using an ECRIS of latest design, we bombarded a clean polycrystalline tungsten target with multiply charged ions of N (q = 4-6), Ne (q = 4-7), Ar (q = 2-12), or Kr (q = 9,11). Ions extracted from the ECRIS have been mass analyzed, transported to a UHV target chamber, decelerated, and focused on the target surface. Target and electron collector currents have been measured at the positive deceleration potential. In this way, total electron emission yields γ_q could be determined down to impact energies of about 10 eV/amu, corresponding to $v = 4 \times 10^4$ m s⁻¹.

Before taking measurements at a background pressure of typically 10^{-7} Pa the target surface was cleaned by Ar⁺ sputtering. The absence of surface contamination was frequently checked by reference to the wellestablished electron yield for Ar⁺ impact on atomically clean tungsten.^{6,12} Results for γ_q obtained with Ar^{q+} up to q = 12 are shown versus impact velocity v in Figs. 1 and 2. Reproducibility of the data is demonstrated by the error bars. The absolute errors increase from 10% for low q up to about 20% for the highest q. We note the initially decreasing electron yields towards a minimum with increasing v, with this effect being more pronounced for the higher q, where the minimum is also shifted toward higher v.

From data for q=2 the contribution due to kinetic emission has been estimated by assuming KE to cause the increase of γ_2 beyond its minimum (compare the shaded area in Fig. 1). Assuming further that $\gamma_{\rm KE}$ is independent of q, the resulting correction becomes relatively small for higher q.

In Fig. 3 the γ values for all measured ion species and charge states at low impact velocity ($v = 0.4 \times 10^5 \text{ m s}^{-1}$) have been plotted versus ion potential energy W_q , while corresponding data for $v = 2.0 \times 10^5 \text{ m s}^{-1}$ are shown in Fig. 4. One clearly observes at low velocity the linear proportionality between W_q and γ to hold for all ions and charge states, whereas at higher v the γ of the higher charge states drop off from the indicated linear relationship. To show in more detail the characteristic impact velocity dependence, data for Ar^{q+} ions with three different impact velocities have been plotted versus ion potential energy W_q in Fig. 5.

All ionization potentials $I_{i-1,i}$ have been taken from Ref. 13. For ions up to $\operatorname{Ar}^{9+}(W_q \approx 1 \text{ keV})$ an almost linear dependence corresponding to relation (1) is apparent, with the factor k, however, decreasing gradually for higher v. In addition, for q > 9 the data for $\gamma_{q,\text{PE}}$ start to fall short of the predictions of relation (1), except at the lowest impact velocity, this effect becoming more pronounced when increasing q and/or v. With the experimental evidence obtained so far, we may explain the emis-



FIG. 1. Total electron yields γ for impact of Ar^{q+} on clean tungsten vs impact velocity v. Shaded area below data for q=2 indicates estimated contribution from kinetic emission, which is believed to be independent on q.



FIG. 2. Total electron yields γ for impact of Ar^{q+} on clean tungsten vs impact velocity v. For q=9, 11, and 12 dashed lines have been added for clarity.

sion of electrons due to bombardment of a metal with relatively slow multiply charged ions as follows.

During approach of an ion Z^{q+} toward the surface, a large number of consecutive neutralization steps will take place, each one probably triggered by (multiple) resonant electron capture from the surface valence band, with subsequent AI and/or AD processes giving rise to emission of electrons. As argued in Ref. 2, each of these deexcitation steps should relax the excited projectile by a small energy amount W of typically 15–30 eV because for larger steps the Auger transitions become rapidly less probable, and for smaller ones the escape probability of electrons from the solid rapidly drops off. Direct experimental evidence for this behavior has been provided from kinetic energy distributions of ejected electrons, which in general seem not to extend beyond about 30 eV.^{3,6,14}



FIG. 3. Total electron yields γ for impact of various ions on clean tungsten vs total ion potential energy W_q at a given impact velocity of 0.4×10^5 m s⁻¹.



FIG. 4. Total electron yields γ for impact of various ions on clean tungsten vs total ion potential energy at a given impact velocity of 2.0×10^5 m s⁻¹.

To achieve complete neutralization, an ion Z^{q+} with total energy W_q therefore has to undergo a number s_q of neutralization steps with

$$s_q \equiv W_q / W . \tag{2}$$

In the course of this simple discussion we neglect all details of the surface as its density-of-states distribution, work function ϕ , etc., which to first approximation are taken care of by the empirical factor k in relation (1), as long as $s_q \gg 1$.

As an example for highly charged projectiles, let us consider the case of Ar^{12+} , for which $W_{12} \approx 2650$ eV and thus $s_{12} \ge 100$. The first RN process involves electron transfer from near the Fermi level of the target metal into



FIG. 5. Dependence of total electron yield γ on total potential energy of primary ions Ar^q + (q = 2-12) for different impact velocities. Note, that for high q the linear relationship between γ and W_q breaks down.

a highly excited Rydberg state n_q of Ar^{11+} , which can be treated in hydrogenlike approximation as

$$n_q = (q^2/2E_b)^{1/2}$$
$$\implies n_q \propto q . \tag{3}$$

The binding energy E_b of this highly excited state with principal quantum number n_q is about equal to the work function ϕ of the target. For Ar¹²⁺ and $E_b \approx 0.18$ a.u. we obtain from relation (3) $n_{12} \approx 20$. A given n_q corresponds to a classical electron orbit with radius $r_{n,q}$

$$r_{n,q} = n_q^2 a_0 / q$$

$$\implies r_{n,q} \propto q , \qquad (4)$$

which for Ar^{12+} and $n_q = 20$ becomes about 18 Å.

Resonance interactions of slow Na atoms in high Rydberg states with a metal surface have been shown to proceed already at distances d_q of several classical electron orbit radii $(d_q \le 5r_{n,q})$.¹⁵ Applying this experience for approach of slow Ar^{12+} toward the surface, the first deexcitation steps will not occur further away than about 100 Å, a distance which at $v = 10^5$ m s⁻¹ is passed in about 10^{-13} s.

On the other hand, for each RN+AI or RN+AD step a time τ of at least 10^{-15} s is necessary, which shows that the ion-impact velocity quite critically decides on whether complete neutralization can be achieved before the ions touch the surface. Neutralization will go on after the ion has reached the surface, but then the escape probability of further produced electrons will have dropped sharply. Since d_q is proportional to q, [see relation (4)], but $I_{q-1,q}$ is about proportional to q^2 for a given projectile species, the total potential energy will increase more rapidly with q than the available distance within which complete neutralization can be carried out. Therefore, at a given impact velocity, for higher q a progressive deterioration of the efficiency for electron production due to the PE processes is to be expected. In the above estimates, the upward shift of ion states due to the image potential has been neglected because it does not change significantly our conclusions.

In conclusion, the following behavior of the total electron yield γ has to be expected. If

$$d_q / (v\tau) \ge s_q = W_q / W , \qquad (5)$$

complete ion neutralization can be achieved before the surface is reached. However, with increasing v the loci for electron ejection due to the consecutive deexcitation steps will gradually shift nearer to the surface, with a correspondingly dropping efficiency for electron emission.¹ This is apparent from our results in Fig. 5, where the factor k in the linear relationship (up to q = 9) slightly decreases toward higher impact velocity. As a further cause, the resonance neutralization probability can be expected to decrease with increasing v.

However, if q and thus W_q become too large for relation (5) to hold, the PE efficiency should progressively de-

crease toward higher q and also v, then causing a significant deviation from relation (1).

Since the first neutralization $Z^{q+} \rightarrow Z^{(q-1)+}$ consumes the relatively largest part of the total neutralization time $s_q \tau$, for higher q and v ions $Z^{(q-1)+}$ can quite probably reach the surface, where they can undergo close encounters with target atoms. Consequently, for impact of highly charged ions, inner shell vacancies resulting in Auger electron emission can be produced at impact velocities, which would be too low to cause such effects for the low q ions.¹⁶

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