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Threshold of ion-induced kinetic electron emission from a clean metal surface

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Electron emission from clean gold under impact of H^+ or singly charged rare-gas ions in their ground state has been investigated at the kinetic emission threshold, measuring both the emission statistics and total yields by means of counting techniques. As we are able to subtract any contribution from potential emission, we observe kinetic emission well below the threshold for sufficient energy transfer from projectiles onto quasifree metal electrons and relate this contribution to quasimolecular autoionization in close collisions between the neutralized projectiles and metal ion cores.

I. INTRODUCTION

Electron emission as a consequence of slow ion impact on a clean metal surface is commonly ascribed to two different mechanisms, the potential emission (PE) and kinetic emission (KE) processes, respectively. PE results from Auger-type electronic transitions involving metal conduction-band states and the initially vacant projectile states.¹ For singly charged ground-state ions it is only observed if the ion potential energy (i.e., the first ionization potential of the corresponding neutral atom V_i) exceeds twice the metal surface work function W_{Φ} . PE is not subject to an ion-impact energy threshold as the KE process, which transfers projectile kinetic energy onto electrons in the metal to eject them above the surface barrier into vacuum.² For clean metal surfaces the KE threshold energy is clearly observable³ and should be ascribed to that projectile energy, where the largest possible energy transfer onto a metal electron just equals the surface work function.⁴

To a first approximation the metal electrons (mass m_e) may be considered as freely moving with velocities of up to the Fermi velocity v_F . Then the maximum energy transfer ΔE from a heavy projectile ion (velocity v: $M \gg m_e$) in a head-on collision is given by⁴

$$\Delta E = 2m_e v \left(v + v_F \right) \tag{1}$$

and, if the electron in a second collision reverses its direction, the KE threshold velocity v_{th} is obtained for ΔE becoming equal to the surface work function W_{Φ}

$$v_{\rm th} = -\frac{v_F}{2} + \left(\frac{v_F^2}{4} + \frac{W_{\Phi}}{2m_e}\right)^{1/2}.$$
 (2)

For a clean polycrystalline gold surface ($W_{\Phi} \approx 5.1 \text{ eV}$, $v_F \approx 1.58 \times 10^6 \text{ ms}^{-1}$, cf. Ref. 5), Eq. (2) thus yields a KE threshold energy of about 315 eV/amu. However, this value is an upper limit, because metal valence electrons may exchange momentum with the crystal lattice. This can be taken into account by ascribing an "effective mass" $m_e^* > m_e$ to the electrons at the Fermi edge. Data in Ref. 6 indicate that for Au this effective electron mass differs only slightly from $m_e(m_e^* \approx 1.09m_e)$, thus decreasing the threshold impact energy from 315 to about 270 eV/amu.

However, recent investigations for impact of Xe⁺ on

clean polycrystalline gold⁷ have revealed a considerably smaller v_{th} than the above given "conventional" value. This could not be explained by PE, because the observed electron yield is continuously falling with decreasing impact energy. Unfortunately, systematic investigations of KE threshold behavior are not only impeded by the rapidly decreasing emission yields γ , but also by so far unseparable contributions from PE, which may also result from long-lived excited ion-beam fractions.⁸ Furthermore, there is a strong dependence of γ on the surface conditions.

In this work we have investigated the KE threshold behavior for impact of slow ground-state ions on atomically clean gold, making use of counting techniques for measuring both total yields down to $\gamma \ge 10^{-4}$ and the emission statistics.⁹

II. EXPERIMENTAL METHODS

Singly charged ions have been accelerated to 4 keV plus the desired final impact energy (cf. below), focused by a magnetic quadrupole doublet, charge-to-mass analyzed and directed into a differentially pumped UHV chamber (base pressure during measurements of approximately 10^{-10} mbar). A four-cylinder lens is used for ion refocussing and deceleration to the desired impact energy on the target surface. Figure 1 shows the last lens element with subsequent parts at potentials for a final ion impact energy of 100 eV. The target consisted of sputter-cleaned high-purity polycrystalline gold (for cleaning procedures cf. Ref. 9). It was placed inside a conical, 96% transparent electrode and a three-electrode cylinder lens assembly, by which the emitted electrons could be deflected and accelerated into a solid-state detector (Canberra passivated implanted planar silicon detector PIPS 100-12-100) biased at 26 kV with respect to the target. For electron counting (mode I operation) the conical electrode was biased at -60 V with respect to the target, as indicated in Fig. 1. Essentially all electrons ejected from the target surface with energies of ≤ 50 eV into a solid angle of 2π reach the active detector surface, as could be proven by methods described in Ref. 9.

Direct measurement of the electron emission yield (mode II operation) was carried out by turning off the detector high voltage and setting the conical electrode al-

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FIG. 1. Experimental setup for measuring electron emission yields and statistics.

ternatively to about +100 or -100 V with respect to the target, thus assuring either complete collection or reflection of the emitted electrons. In this way, the total electron yield γ can be simply derived from

$$\gamma = \frac{I^+ - I^-}{I^-},$$
 (3)

where I^+ and I^- are the target currents measured for positive and negative voltage at the conical electrode, respectively. Mode II measurements of γ required ion fluxes of $\geq 10^{11}$ s⁻¹ and could only be carried out with sufficiently large differences between I^+ and I^- (i.e., for $\gamma \geq 0.05$), whereas for still smaller γ we applied the counting mode I in the following manner.

The original ion beam was attenuated in a well-defined, reproducible way by detuning the current for one element of the magnetic quadrupole doublet, which procedure did not interfere with the ion deceleration in front of the target. Therefore, the attenuation factor was independent of the primary ion beam intensity and/or final impact energy. It could be precisely determined at ion impact energies, where γ was still conveniently measurable via mode II experiments. Considering the electron emission statistics (ES)⁹

$$\gamma \equiv \sum_{n=1}^{\infty} n W_n, \quad \sum_{n=0}^{\infty} W_n \equiv 1$$
(4)

 $(W_n$ is the probability for emission of *n* electrons per primary ion), and Eq. (3), we obtain the following condition:

$$\gamma I_i \equiv I_e = \sum_{n=1}^{\infty} neC_n \,. \tag{5}$$

 C_n are the measured electron count rates for different *n*, which according to Eq. (4) are related to the probabilities W_n by

$$C_n = \frac{I_i}{e} W_n \tag{6}$$

and, therefore, can deliver I_e via Eq. (5). By already knowing γ from mode II measurements, we could then evaluate the attenuated I_i and thus the corresponding attenuation factor. The mode I procedure could deliver precise electron yields γ at still lower impact energies, where the mode II measurements were no more applicable. In this way we could measure yields as low as 2.5×10^{-4} electrons per ion, with an accuracy mainly depending on the amount of particle reflection from the target surface. In the investigated low-impact energy range ($\geq 100 \text{ eV}$), reflection coefficients for heavy projectiles on a clean gold surface become typically 10-50%.¹⁰ Although most of the reflected projectiles are neutral and therefore cannot disturb the measurement of I_i , they cause emission of electrons at the conical electrode. These electrons are also collected by the detector, thus causing a systematic error which could be estimated from published coefficients for particle reflection on clean metal surfaces,¹⁰ the 96% transparency of the conical electrode, and γ values for impact of rare-gas ions on gas-covered tungsten¹¹ as the material of the conical electrode.

To avoid long-lived excited ion-beam fractions, our plasma ion source has been operated at sufficiently "weak" discharge conditions.¹²

III. RESULTS AND DISCUSSION

Figures 2(a) and 3(a) show total electron emission yields γ measured in mode I (calibrated to yields $\gamma > 0.05$ obtained from mode II measurements, cf. section II) at impact energies near and below the "conventional" kinetic emission threshold.⁴ The error bars include all systematic and statistical contributions. With the exception of Xe⁺ $(V_i = 12.1 \text{ eV vs } W_{\phi} \approx 5.1 \text{ eV for Au})$ we clearly observed PE contributions for all ion species.

At the lowest accessible impact energy (E = 100 eV)the apparent magnitude of γ_{PE} was in fair agreement with semiempirical predictions,¹³ indicated by dotted lines on the left-hand side of Figs. 2(a) and 3(a). Moreover, for He⁺ impact Fig. 2(b) shows the important result, that the value W_2/W_1 (i.e., the ratio of probabilities for emission of, respectively, two and one electrons) approaches zero (corresponding to $W_2/W_1 < 1 \times 10^{-2}$ as the detection limit), if the region of apparently exclusive PE is entered. The same was found for impact of Ne⁺ and Ar⁺ [cf. Fig. 3(b)] and should hold as well for any slow singly charged ground-state ions impinging on gold. We, therefore, conclude that in the PE process at most one electron can be emitted, which can be explained by the rather small correlation of gold surface states at the Fermi level. Consequently, the appearance of a second electron (i.e., W_2/W_1 > 0) has to be attributed to the onset of KE, which then



FIG. 2. (a) Total electron emission yields for H^+ (open circles) and He^+ (solid circles) impact on clean polycrystalline gold vs impact energy per atomic mass unit. The vertical arrow indicates the "conventional" threshold for kinetic electron emission as calculated from Eq. (2). Semiempirical predictions (Ref. 13) for the potential emission yields are indicated at the left-hand side by dashed horizontal lines. (b) Ratio of emission probabilities W_2/W_1 for H^+ (open circles) and He^+ (solid circles). Vertical arrows as for Fig. 2(a).

clearly determines the KE threshold also in cases where the total yield is dominated by PE contributions.

For H⁺, He⁺, Ne⁺, Ar⁺, and Xe⁺ impact on gold, significant KE contributions (i.e., $\gamma_{KE} \ge 1 \times 10^{-2}$ electrons/ion) were still found down to 150, 80, 20, 10, and 10 eV/amu, respectively.

For Xe⁺ our γ measurements agree well with Ref. 7 down to $E \approx 10$ eV/amu. The apparent discrepancy at lower impact energy is related to the application of different experimental techniques, with our method being probably more reliable.

Obviously, all projectile ion species cause nonvanishing KE contributions well below the conventional threshold derived from Eq. (2). This we explain by quasimolecular autoionization¹⁴ in close encounters of the neutralized projectiles with Au⁺ target ion cores, as already speculated in Ref. 4. A clear support for this conclusion is the fact that the KE yields below the conventional threshold become relatively larger for the heavier projectile ions, as seen from a comparison of Figs. 2(b) and 3(b). We mention that for gas-covered metal surfaces several groups have observed¹⁵ KE yields, which at given impact energy are slightly higher for slow neutral particles than for the corresponding singly charged ions. This can be related to a higher probability for electron emission in close en-



FIG. 3. (a) Total electron emission yields for Ne⁺ (open squares), Ar⁺ (open circles), and Xe⁺ (solid circles) impact on clean polycrystalline gold vs impact energy per atomic mass unit. Crosses show results for Xe⁺ from Ref. 7. Vertical arrows and dashed horizontal lines as for Fig. 2(a). (b) Ratio of emission probabilities W_2/W_1 for emitting, respectively, two and one electrons for impact of Ne⁺ (open squares), Ar⁺ (open circles), and Xe⁺ (solid circles) on clean polycrystalline gold vs impact energy per atomic mass unit. Vertical arrows as for Fig. 2(a).

counters of the neutral species with surface adsorbates, taking also into account the incomplete ion neutralization at a contaminated metal surface.

IV. CONCLUSIONS

We have measured total yields for slow ion-induced electron emission from a clean gold surface. An unambiguous separation of the contributions from, respectively, potential and kinetic emission could be achieved by considering the also measured electron emission statistics. It is shown that for impact of singly charged ground-state ions PE can produce, at most, one electron. For the KE threshold upper limits could be derived, which are inconsistent with a direct projectile energy transfer onto the valence electrons. It is, therefore, concluded that below the conventional KE threshold ion impact energy quasimolecular autoionization in close encounters between the neutralized projectiles and target-ion cores becomes the dominant source for kinetic electron emission.

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