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Ejection of Electrons from Molybdenum and Tungsten by Kilo-Electron-Volt Potassium Ions*

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The experimental coefficient of ion-electron emission for K^+ in the 0.6–6-keV energy range incident on pure Mo has been found to be in excellent qualitative and numerical agreement with the calculation by Parilis and Kishinevski. Sharp thresholds were measured at, respectively, 0.59 and 0.64×10^7 cm/sec for Mo and W. Evidence involving the distribution in energy of electrons ejected at angles near the target normal showed that incident-beam particles embedded in the target fundamentally affect the nature of the ejection process. This contamination factor is basically the cause of the discrepancy between the new and previous data. In particular, the linear intercept for Mo differs by 50% from Brunnée's value. Pulse measurement techniques were used and pure Mo surfaces were prepared by subliming away contaminated metal layers at 2000 °K. The electron emission for K^+ on Mo was found to be augmented by monolayer coverage of pure Mo by N_2 at low energies but was reduced slightly at incident energies above 3 keV. This observation is discussed in terms of the surface-barrier effect on escaping electrons.

I. INTRODUCTION

Past studies of the secondary-electron emission by bombardment of assumed "atomically clean" metal surfaces with alkali ions of ionization potential less than the surface work function (kinetic emission) are in several important cases contradictory. This investigation set out to verify the yield data for $K^+ \rightarrow$ Mo by Brunnée[1] and $K^+ \rightarrow$ W by Petrov,[2] which are of particular interest since these pairs and Rb^+ (or Kr^+) on Mo and W were specifically treated in the calculation of kinetic emission yield by Parilis and Kishinevski,[3] henceforth referred to as PK. These earlier experimental results are significantly at variance to the theory. However, a discrepancy exists between present and prior data that is assumed to be due to target contamination by incident particles under earlier experimental procedures. In the current work, obvious beam-contamination effects were eliminated by reducing the beam current until good reproducibility was established. Under these conditions, the yield curves of Brunnée and Petrov were reproduced quite well; but as discussed in detail in Sec. III A, concrete evidence accumulated that despite this deceptive reproducibility, contamination effects yet occurred. Thus, the data reported here were taken by means of single-current pulses of several msec each. In the case of Mo, layers contaminated by embedded particles were eliminated periodically by subliming away ≈100 atomic layers of the target surface at 2000 °K. Influence of contamination then was eliminated from the Mo data and minimized in the W results. Under these conditions, sharp thresholds were measured in each case, whereas previously these had been obscured. It was also found that the linear intercepts were greater as the yield was lower.

The experimental yield curves are presented in Sec. III B and compared to the PK calculation. Section III C presents data for yield when K^+ is incident on pure Mo covered by a monolayer of N_2, and the effect of gas coverage at high and low ion energies is discussed. Also, it is suggested that the similarity of an undulation in present Mo/N_2 data and "atomically clean" yield data by Magnuson and Carleston[4] might mean that, in fact, the "atomically clean" supposition was incorrect.

II. APPARATUS AND PROCEDURE

A. Experimental Tube

Figure 1 shows a schematic diagram of the operating portions of the apparatus. The complete

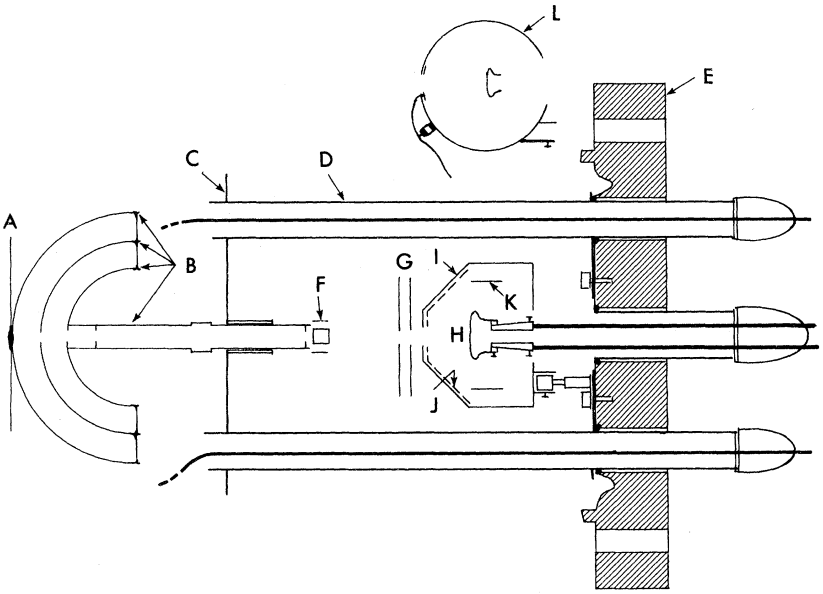

FIG. 1. Schematic diagram of the flange-mounted apparatus. The individual components are as follows: A, coated-filament ion source; B, ion-gun electrodes (held in place by sealed-on glass); C, stainless-steel disk; D, stainless-steel tubes (4 altogether); E, stainless-steel flange, highly schematic; F, deflection plates; G, collimators; H, foil target; I, Faraday cage; J, grid; K, secondary-electron collector; L, alternate electron collector used for RPD analysis.

assembly was mounted on a 4-in.-i.d. 304-stainless-steel flange. The structural layout provided good shielding of the target region from the high-voltage leads and electrodes. The pure source of K^+ ions was a resistance-heated iridium filament, coated with an alumino-silicate molecular sieve prepared in the manner described by Weber and Cordes.[5] The beam was tested for purity by means of a time-of-flight mass spectrometer with the apparatus in its normal operating mode except that the target had been replaced by an electron multiplier. The K^+ beam showed no impurity to the level of 0.5%. The second of the hemispherical ion accelerating electrodes was *RC* coupled to a pulse generator to form 1- to 10-msec pulses where needed. Beam current density at the target was measured with some care and found to be roughly uniform over a diameter of 1.5 mm and to cut off sharply; i.e., less than a fraction of 10^{-3} of the beam current was intercepted at the collimator (G) of Fig. 1. The targets were of very pure Mo or W foil, 0.01-mm thick, and had an area of 5×6 mm^2 normal to the incident beam. Molybdenum lugs attached to tungsten leadouts secured the filament. Metal-to-ceramic-to-metal seals, normally used as feedthroughs, were used as insulating supports for the collector, semifinal collimator, and deflection plates.

B. Electron Collection

Some care is required in the collection of electrons ejected by alkali ions incident on clean metal surfaces. The problem is one of separating away, or correcting for, the reflected ion current and the considerable current of tertiary electrons liberated when reflected ions strike the collector walls. The present approach was to separate the currents physically. The electron collector for total yield (Fig. 1) was symmetric around the beam axis. A hoop-shaped electrode (K) was placed flush with the target plane to collect the target electrons. Reflected ions could not reach this hoop as it was behind the plane of the target, and tertiary electrons liberated at the collector walls were returned by a negative-biased grid (J). A positive bias of 160 V was applied to the hoop collector which, being intentionally unshielded by the screen, could establish an extracting field at all points on the target surface for electrons, while the screen was biased to -25 V to turn faster electrons emitted near the target normal back to (K).

These arrangements were carefully tested as follows. Thermionic electrons from the target were collected by the hoop (K) to the extent of $100\pm2\%$. To test for collection of the higher-energy emitted electrons, these were produced by bombarding a gas-covered target by 8-keV incident ions. Here, collection conditions were more favorable so that the yield could be measured directly with the collector shell and the suppressor grid, and a reasonable correction could be made for reflected ions. This test indicated a 100% efficiency with perhaps 3% uncertainty owing to possible wrong estimates of the reflected-positive-ion energy spectrum. Finally, it was found that measured yield under experimental conditions was unchanged by variations of the bias voltages of about 20%.

The collector shell of 50-mm diam was constructed of stainless steel, the screen was electroformed nickel of 90% transparency, and the hoop was formed from 7-mm-wide Ta sheet. The geometry of this collector shell was rather arbitrary

and chosen for convenience of construction. For the energy spectrum analysis, however, a spherical collector (L of Fig. 1) of 50-mm diam was required so that the usual retarding-potential-difference (RPD) method might be applied. The optional insert was used to capture electrons emitted within 18° of the surface normal. The magnetic field in the target region was $\lesssim 0.1$ G and not troublesome. However, this geometry suffers from the focusing effect caused by a potential difference between the sphere and the insert electrode.

Additional details concerning the apparatus and procedure are available from Ref. 6.

C. Data Recording

Ion-beam voltage at each measurement point was known to 0.5% accuracy including a 2-V drop across the filament source. Two currents were measured: (i) collection-electrode current and (ii) current to the screen, collector shell, and target. Since the absolute value of current was not required, good accuracy was obtained by cross calibrating the recording instruments. For pulse measurements, the currents were fed to a dual-trace oscilloscope except that a preamplifier with 10^7-Ω input impedance was used to measure the reduced electron current near threshold. Calibration was accurate to better than 2% but a random error was present in the form of noise to about 3%. In the average of eight or so measurements, this meant a net possible error of, say, 4% or less in yield magnitude. In practice, resolution near threshold was typically $\pm 5\times10^{-4}$.

Electron energy distributions were also measured. In order to form the spectral curves, linear variation of the retarding potential in time and *RC* differentiation of collector current were used and the result of each 10-sec scan was recorded by a chart recorder.

D. Vacuum System and Procedure

The design of the vacuum system and ultrahigh-vacuum (uhv) technique differed little from the standard Alpert techniques of previous experiments at this laboratory.[7] The vacuum envelope was mainly of 4-in.-diam stainless-steel tubing which sealed via a Kovar-7052–glass graded seal to a glass-mercury-diffusion-pump system with liquid-N_2 traps. A titanium sublimation pump, not used in the earlier experimental systems, was added and used to speed processing to uhv.

With the aid of sublimation pumping, the pressure prior to bakeout was reduced to less than 10^{-6} Torr. At this time, the filaments were outgassed at or above their operating temperatures for 2 h. A 300 °C 24-h bake of the entire system, including the traps, followed; after this, the base pressure was typically on the order of 5×10^{-10} Torr if the system had been "up to air" for only an hour or so, as was generally the case. It was then necessary to heat the filaments at operating temperatures for 24 h to outgas them to uhv and then, typically, the pressure was $1-2\times10^{-9}$ Torr with the filaments in steady operation. Sublimation pumping was also useful during this outgassing period.

Target-surface cleaning was by the conventional flash-filament technique. The Mo- and W-foil targets were resistance heated to temperatures of 1750 and 2000 °K, respectively, for about 1 day in uhv. Subsequent surface purity was maintained by flashes (2200 °K for W) of a few sec at regular intervals with $p\lesssim 1\times10^{-9}$ Torr. As mentioned in Sec. III A, it was found necessary to take yield measurements only with single ≈ 5-msec pulses of typically 2×10^{-8}-A peak current. Transformed to a flux density and referred to the surface density of metal atoms, this was $\approx 10^{-2}$ monolayers/sec or $\approx 10^{-4}$ monolayers/pulse. During experimentation with Mo, after every 20 or 30 current pulses, approximately 50 atomic layers were evaporated away to eliminate entrapped K atoms. As this procedure was not practical for W, the total number of pulses was limited to roughly 100 after which the experiment was terminated. Two W targets were studied in this way.

Additionally, yield was measured for monolayer N_2 coverage of clean Mo. For this purpose, the Mo target was flash cleaned with $p\lesssim 1\times10^{-9}$ Torr and then nitrogen was admitted to a pressure of 5×10^{-8} or 5×10^{-7} Torr. About five measurements by single 2-msec pulses were performed beginning either ≈ 1 min or ≈ 10 sec after admitting the gas, depending on the pressure. Since adsorption of a second gas layer proceeds at a rate reduced by more than a factor of 10,[8] it is possible to conclude that these measurements were associated with neither significantly more nor less than monolayer coverage. That no variation occurred in each succession of five measurements further supports this. However, it was found that this measurement is even more strongly influenced by alkali contamination than the clean-surface measurement!

III. RESULTS AND DISCUSSION

A. Alkali Contamination of a Mo Target

The phenomenon of surface contamination mentioned in Sec. I bears amplification. At the beginning of the investigation, it was found that electron yield and ion reflection increased by as much as 100% during periods of bombardment of 100 sec by a K^+ particle flux of 10^{13} $(cm^2\,sec)^{-1}$. The number of particles embedded by past bombardment was seen to be a variable in that the initial value after a flash was not reproducible. A natural remedy was to reduce the incident-particle flux density to $\lesssim 10^{12}$ $(cm^2\,sec)^{-1}$, limit the current measurements to $\lesssim 10$ sec, and heat the Mo filament to the outgas-

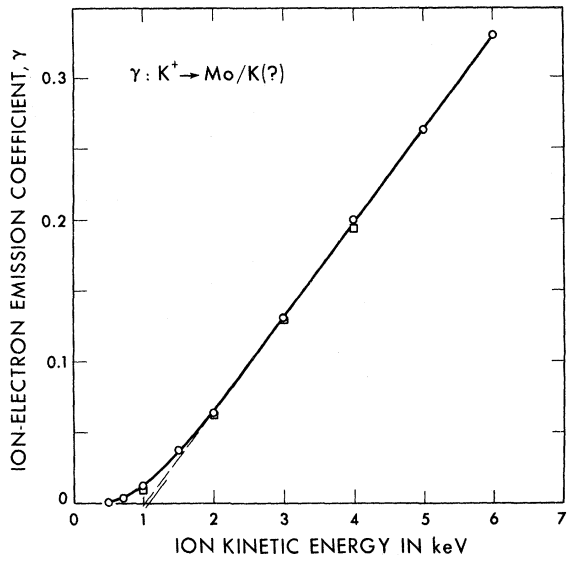

FIG. 2. Variation of γ with ion energy for K^+ incident on conventionally cleaned Mo. Data points, given as squares, are from the work by Brunnée (see Ref. 1).

sing temperature, 1750 °K, for ≈15 h after a number of measurements, say 20. This caused electron yield and reflection probability to be constant during the measurement period, and the reproducibility of these values was provisionally accepted as proof that contamination effects had been eliminated. This is the conventional technique apparently used by Brunnée, [1] Waters, [7] and others.

Figure 2 shows the values of the secondary-emission coefficient γ for K^+ ions normally incident on conventionally cleaned Mo, given as circles compared to the squares reported by Brunnée. The linear intercepts of the two sets of data agree to 5%. The yield behavior in Fig. 2, below 1 keV, is equivalent to the finding of Waters for Li^+, $Cs^+ \rightarrow W$, but, as Waters also found, the relative scatter of the data increased without apparent cause for kinetic energies below about 1 keV.

This last inconsistency and studies of the electron energy distributions led to the discovery of a more subtle or second degree of contamination, which might be called β contamination, by embedded particles. Figure 3 gives the measured spectrum of electrons ejected from Mo by K^+ incident and this result agrees with that of Brunnée except the absolute cutoff indicated at ≈13 eV was not confirmed. Then, the energy spectrum of electrons ejected within 18° of the target normal was measured and this result is the "leftmost" solid curve of Fig. 3. The distribution appeared to peak, and this reproducibly, at 0 eV which is at considerable variance to the theoretical expectation. On account of refraction and internal reflection of emerging electron waves at the metal surface, the escape probability should rise from zero at zero electron energy relative to the vacuum level. Since the argument applies to electrons ejected at angles near the target normal as well as to the total distribution which clearly shows the expected effect, this result was certainly anomalous.

In order to eliminate all entrapped K particles which might cause what appeared to be an anomalous surface interaction, ≈ 100 atomic layers of the metal target were "evaporated" away at a temperature

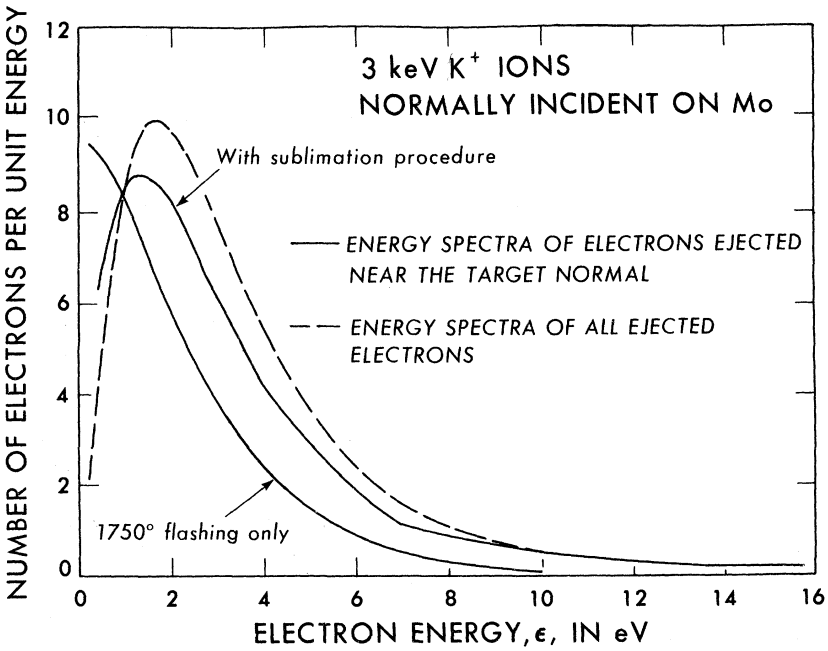

FIG. 3. Distribution in energy of electrons ejected from Mo under the former and new experimental techniques. While the total distribution was approximately unchanged, the energy distribution at angles near the surface normal showed a considerable difference. The partial distributions are multiplied by 10.

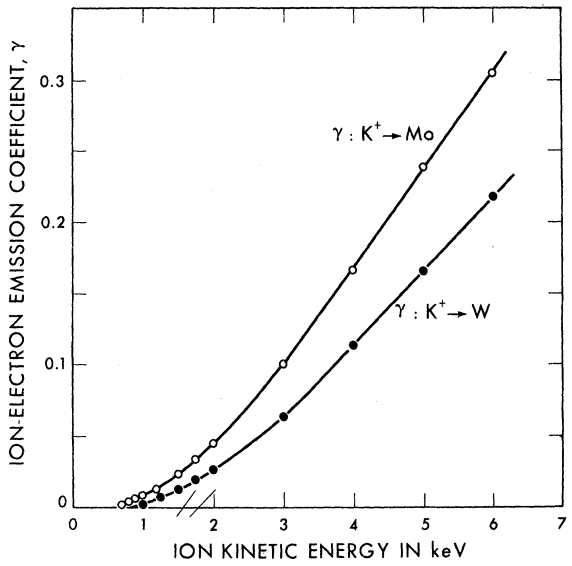

FIG. 4. Variation of γ with ion energy for K^+ normally incident on pure Mo and W.

of 2000 °K. Here, tabulated evaporation and electron-emission values were used to estimate the mass sublimed away. This showed that impurity atoms in the surface layers caused the anomaly since the normal curve of Fig. 3 was subsequently recorded. Additionally, when yield was measured with single 10-msec pulses only, yield for a "virgin" target was found to be reduced in magnitude from the curve of Fig. 2 to that of Fig. 4. But after ≈30 current pulses, reproducibility deteriorated and measured values tended to drift up to their former values. Since the sublimation procedure consistently restored the initially lowered values, it must be concluded that there had been K-particle contamination when the curve of Fig. 2 was measured and, in general, when the conventional experimental technique had been applied. This, then, is a second and more subtle effect that requires relatively few incident particles, that is not cured by the 1750 °K outgassing procedure, and that produces ion-electron emission results of good reproducibility. However, the anomalous distribution for electrons ejected near the target normal, while incidentally the total spectrum was affected to only about 10%, indicates that the effect of β contamination on ion-electron emission is more extensive than a mere shift of the surface work function. The fact that a threshold found for the pure surface (Fig. 4) was totally obscured in the presence of contamination likewise indicates a nontrivial change in the nature of the emission process.

Ironically enough, the outgassing procedure is important in setting up the stable and reproducible β contamination state. Roughly 0.5 monolayers of 3-keV K^+ bombardment followed by 12 h of heating to 1750 °K resulted in the reproducibly anomalous curve of Fig. 3. On the other hand, ten times this number of incident particles were embedded in the target in the presence of only periodic flashes of a few sec each, and yet the measured spectrum curve more closely resembled the normal than the anomalous curve of Fig. 3. However, a measurable effect on yield was first noted when ≈0.01 monolayers of K atoms had been shot at the target.

The nature of the contaminant state is open to speculation. It has the singular characteristic of being reproducible and, in view of the distribution findings, must be due to the presence of alkali ions on or near the surface. Since the larger level of contamination initially observed produces considerably greater yield, it follows that the degree of K coverage must be constant in second-degree contamination. One possibility is that the stable impurity state occurred when the K volume concentration reached its equilibrium level at 1750 °K, and that ions diffused to the surface as the filament cooled after each flashing. Another possibility is that there are surface sites where the binding energy is sufficient to hold a K atom for a period on the order of the reciprocal diffusion rate of K atoms to such surface sites when the metal is heated to 1750 °K. It is plausible that the fraction of sites with this property might be constant under flash-filament processing.

B. Yield Data for K^+ Incident on Clean Mo and W Surfaces

The measured electron yield from atomically clean Mo and W surfaces plotted versus kinetic energy of the normally incident K^+ ions is shown in Figs. 4 and 5. Data points are the average of eight or more measurements each with typically two values discarded. Sublimation cleaning of the Mo target of K ions was performed periodically; but as this was not practical for W, these data are perhaps marginally influenced by K contamination. The Mo and W thresholds are 0.70 and 0.82 keV and the linear intercepts are 1.6 and 1.8 keV, respectively.

Brunnée's results for the $K^+ \rightarrow$ Mo experiments were reproduced using his procedures which were found on analysis to cause contamination of the target surface by beam particles (Sec. III A). Thus, the discrepancy between the linear intercept reported here, 1.6 keV, and that found by Brunnée, 1.05 keV, is most probably due to alkali contamination of the target in the former experiment. The situation as to the earlier $K^+ \rightarrow$ W yield data by Petrov[2] is similar except that the effect on yield is less severe for W. The important point here is that a sharp threshold was found below 1 keV. That Petrov reported a threshold at the linear intercept (1.6 keV) was possibly due to the limitations of the

oscillographic measurement technique near threshold. It is of interest that subsequent to his original investigation, Petrov studied yield for a K^+ beam incident at 45° to a W surface.[9] At this time, more care was given to minimizing alkali contamination and beam current was switched on only during yield measurements, although in periods of seconds rather than the msec used in this work. The linear intercept reported for this investigation was 1.8 keV and since it is believed (see for example, Ref. 3) that target inclination increases yield by a constant factor of $\sec\theta$, this result is in agreement with present data as to the linear intercept. At lower ion energies, an exponential rise was found with considerable scatter in succeeding data points, much as had been the case here when Mo was studied by means of dc measurement techniques.

The form of the yield curves follows closely that predicted by the PK calculation[3]; a mild linear, or less than linear, energy dependence at energies immediately above threshold and linearity at energies above ≈2 keV. As to the PK analysis, the following paragraph will show that while the magnitude of the yield is not particularly suited to experimental comparison, it is useful to compare the computed threshold and linear intercept to the experimental data.

The complete PK yield formula is as follows:

$$\gamma = \int_0^{x_m} \sigma(E)\, w e^{-x/\lambda} N\, dx\,, \tag{1}$$

$$\sigma(E) = \int_0^{s_{\max}} \frac{\epsilon(E, s)}{J}\, 2\pi s\, ds\,, \tag{2}$$

$$E = E(z_1, z_2, M_1, M_2, x)\,. \tag{3}$$

It was assumed that after metal-core atoms are ionized by the impinging ions, conduction-band electrons are excited in the two-electron Auger neutralization of the hole states and these are the electrons that escape the metal. Thus, σ is the cross section for hole formation computed on the basis of an average energy transfer J, per ionization with the use of the Thomas-Fermi-Firsov[10] formula for the collisional inelastic energy transfer ϵ. Since the ϵ calculation is of a highly statistical nature, no free parameters are introduced by it and, in fact, J, w (Auger neutralization probability), and λ (electron diffusion length) are the only parameters of the yield formula not experimentally well known. Further, w/J occurs as a factor and influences neither the threshold or linear intercept and, additionally, λ does not affect the threshold value. Hence, these two numbers, threshold and intercept, provide a fine experimental test of the applicability of the theory.

In their article, PK reported that the computed ratio of linear intercept to threshold was 2.25. This number agrees strikingly well with the experimental ratios 2.3 for Mo and 2.2 for W. The threshold velocities were said to lie in the range 0.6–0.7×10^7 cm/sec for the various ion-target combinations K^+ or Rb^+ on Mo or W. It is possible to conclude that $K^+ \rightarrow$ Mo and W would correspond to threshold velocities at the upper end of the range, 0.7×10^7 cm/sec, in very reasonable 15% agreement with the present experimental results, 0.59 and 0.64×10^7 cm/sec. However, the threshold was computed by using hole depth relative to the top of the conduction band. These numbers were taken

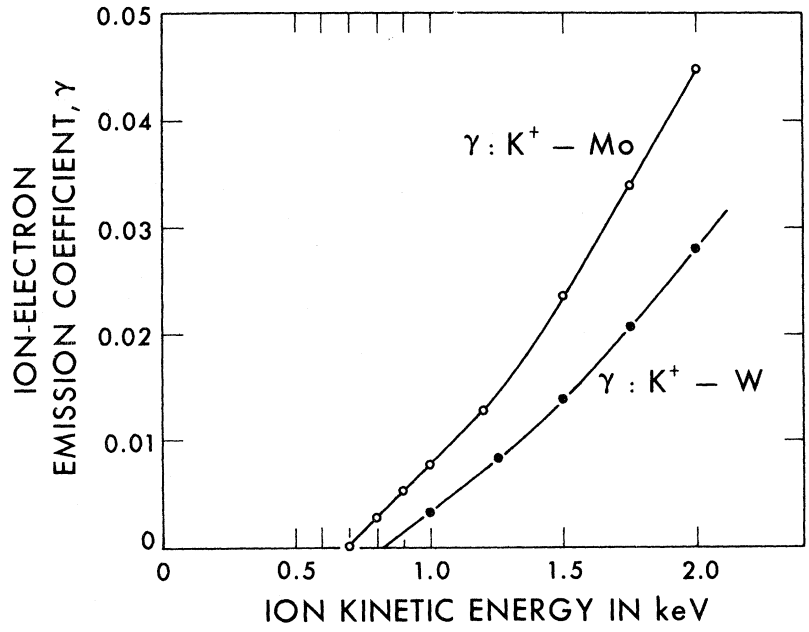

FIG. 5. Variation of γ with energy near threshold for K^+ normally incident on pure Mo and W.

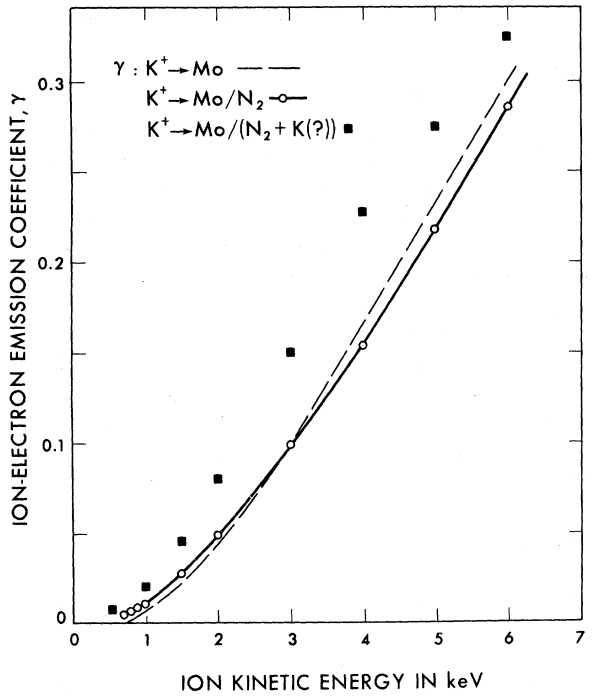

FIG. 6. Variation of γ with ion energy for K^+ normally incident on pure Mo with monolayer coverage of N_2. Also shown is the pure-surface yield curve and yield data points taken under conditions of K contamination (see text).

from inelastic energy-loss data by Harrower[11] to be 11.6 eV (Mo) and 14.8 eV (W), but later and apparently more careful studies by Apholte and Ulmer[12] and by Lynch and Swan[13] quoted values of 10.1 and 9.9 eV (Mo) and 11.1 and 10.3 eV (W). Using an average of these latter numbers and assuming that ϵ is approximately proportional to the square of the ion velocity, the corrected theoretical threshold velocities would be 0.65×10^7 and 0.60×10^7 cm/sec. These are in quite handsome (10%) agreement with the experimental results.

C. Yield Data for Monolayer Coverage of N_2 on Pure Mo

The yield function measured for K^+ incident on a pure Mo surface covered by a monolayer of adsorbed nitrogen gas is in Fig. 6. The individual measurements showed some scatter directly traceable to the presence of beam contamination. To emphasize the severity of this effect, the points enclosed in boxes are shown. For these, after approximately 50 pulses had been directed at the target, the surface was thoroughly outgassed at 1750 °K for 1 h with $p=1\times10^{-9}$ Torr. Then nitrogen was admitted as before and the points depicted by the boxes resulted.

As Waters[7] had reported, yield at 1-keV energies and below are greatly enhanced by monolayer coverage. However, it is natural to question whether the yield obtained in this earlier experiment was increased beyond that for a gas-covered surface by alkali contamination, since a single target was apparently used by Waters for an extended period of time without precaution to remove embedded particles.

Beyond 3 keV, the yield is actually reduced. Particularly since this trend stabilizes, it may be believed that the only effect of monolayer N_2 coverage on the emission process here is to reduce the electron escape probability by increasing the metal work function. The increase in yield below 1 keV is undoubtedly due to electrons "boiled off" in atomic collisions occurring essentially outside the metal surface. But, for incident particles of many kiloelectron-volts, the metal lattice is quite soft and penetrable, to the point that PK could justifiably assume that most fast ion paths are approximately linear within the metal. Thus, at high ion energies, electrons "boiled off" in the K^+-N atomic collision must escape over the surface potential and refractive barrier in order to be observed. From the data, it appears that, in fact, a negligible number of these direct or "boiled off" electrons escape, which is the PK assumption.

These data may also bear on the investigations by Magnuson and Carleston,[4] who conventionally increased the beam density of incident nonreactive noble-gas ions until a leveling off was reached in electron yield. This yield value was taken as that for an atomically clean surface. Arifov *et al.*[14] have already disputed this conclusion as their results for flash-cleaned Mo are not in agreement; also, it may be stated that the discrepancy is most severe in the low-energy region most influenced by contamination. In addition, their yield curves show a characteristic undulation in the neighborhood of 5 keV not found in other clean-surface data. On the other hand, the slope of the curve $K^+\rightarrow Mo/N_2$ declines briefly at about 3.5 keV, as does the $K^+\rightarrow Mo/(N_2+K)$ curve at 4.5 keV in the sort of undulation that is common in the Magnuson and Carleston data. In fact, for their $Ar^+\rightarrow Mo$ (single crystal) curves, there is a slope minimum at 4.5 keV where a tangent would cut the Y axis at about 0.15. Curiously, the $K^+\rightarrow Mo/(N_2+K)$ curve shows a slope minimum at the same point and an intercept at roughly 0.07. The difference here is about the amount of potential emission for Ar^+ on Mo. It would seem possible that their data suffer from not only residual gas contamination but also from contamination by embedded particles of the incident beam. Further, it appears from this that the inert rare-gas atoms embedded in a target may have an effect on the kinetic ion-electron emission process similar in nature to the embedded electropositive alkali atoms.

IV. CONCLUSION

The current results for kinetic ion-electron emission are in excellent quantitative agreement with the results of the PK theoretical calculation most appropriate for numerical comparison, the yield threshold, and extrapolated linear intercept. This is an agreement intrinsically related to the accuracy of the Thomas-Fermi-Firsov statistical model and the PK method of cross-section computation, and credits each.

It is to be concluded that a discrepancy between previous data for K^+ – Mo and W ion-electron yield and the results presented here is due to the presence of beam particles in the targets of the earlier work. It is the author's recommendation, based on this series of experiments, that better precautions be taken than before to ensure against surface contamination in similar experiments. For the cases of flash-cleaned Mo and perhaps W surfaces, a simple means of either removing the embedded particles or verifying that they have no effect is available: A new pure surface may be exposed by subliming away atomic layers which may have an objectionable concentration of incident beam particles.

It has been found that single-layer coverage of clean surfaces does not affect ion-electron emission for sufficiently fast incident ions beyond an amount attributable to a work-function change, and increases yield less at low energies than was previously believed.

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Nuclear-Magnetic-Resonance Analysis of Hydrogen Motion in Hydrides*

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A comparison of the results from analyzing spin-lattice relaxation times (T_1) of diffusing spins using an exponential correlation function and the isotropic model of Torrey has been made. Data from $ScH_{1.7}$, $TiH_{1.55}$, and $TiT_{1.5}$ were used. No significant difference was found for activation energies, the isotope effect, or minimum T_1 using the two models.

I. INTRODUCTION

The general theory of diffusion-induced nuclear relaxation has been fairly well developed.[1] The first formulation of this problem was presented by Bloembergen *et al.*,[2] where only the macroscopic aspects of the phenomena were considered. Later a more complete model was presented by Torrey[3] in which atomic motion was considered as essentially a random-walk problem. Using this approach, Torrey was able to relate, at least for isotropic motion, the pertinent microscopic diffusion parameters to observable nuclear-magnetic-resonance (NMR) properties.