Excited-state formation as H^+ and He^+ ions scatter from metal surfaces*

W. E. Baird, M. Zivitz, and E. W. Thomas
School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332
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Impact of 10- to 30-keV H^+ or He^+ ions on polycrystalline metal surfaces causes some projectiles to be backscattered in a neutral excited state. These projectiles subsequently radiatively decay, emitting Doppler-broadened spectral lines. By analysis of the spectral shape of these lines, we are able to determine the probability of radiationless deexcitation of the excited backscattered atoms. Quantitative measurements of spectral intensity indicate that less than 1% of all projectiles are backscattered in an excited state. The relative variation of total spectral line intensity with angle of projectile incidence and with projectile primary energy has been successfully predicted using a model which assumes that the probability for excited-state formation is independent of the scattered projectile's energy and direction. We also predict the variation in total spectral line intensity with target atomic number. Finally, we examine briefly the sputtering and excitation of Al under He^+ impact.

I. INTRODUCTION

It has been shown[1,2] that when 3- to 30-keV He^+ ions are incident on a metal surface, some of the projectiles are scattered as neutral excited atoms. These atoms subsequently decay by photon emission. The resulting spectral line is Doppler-broadened, and the line shape is directly related to the distribution of speeds and distributions in angle of the scattered projectiles. In a recent paper[2] we described a theoretical model which predicts these line shapes using the backscattering theory of McCracken and Freeman.[3] In order to explain the absence of slow excited atoms, we included a radiationless deexcitation term by which the excited electron may be lost while the recoiling atom is still in the vicinity of the surface; possible deexcitation mechanisms include the Auger effect and resonant ionization.[4] The radiationless deexcitation can be approximately represented by the following expression which gives the probability R that a particle having a velocity component $V_\perp$ perpendicular to the surface of the metal will escape without radiationless decay[4]:

$$R(V_\perp) = \exp(-A/aV_\perp) \ . \tag{1}$$

In our previous work we utilized the analysis of measured line shape to determine the ratio A/a; we refer to this ratio as the "survival coefficient."

The principal objectives of the present work were to perform further measurements of the survival coefficient for excited helium and hydrogen atoms scattered from surfaces, and to study quantitatively the probability of excited-atom formation as a function of projectile energy. We utilized H^+ and He^+ projectiles of energies from 10 to 30 keV incident on polycrystalline targets of aluminum, copper, niobium, molybdenum, silver, and tungsten; spectroscopic observations were made in the visible and near ultraviolet regions. In the case of He^+ ions incident on aluminum, we also observe emissions characteristic of sputtered aluminum atoms; these emission intensities have also been measured and are presented here.

The experimental arrangement was identical to that in our previous work[2] and will not be described in detail here. The He^+ or H^+ ions obtained from an rf discharge source are mass analyzed, collimated, and directed onto the target surface at some incidence angle ϕ with respect to the target-surface normal. A grating monochromator views the surface through a sapphire window; the monochromator axis is perpendicular to the projectile beam direction, and lies in the same plane as the projectile beam and the target-surface normal. The targets were maintained at a base pressure of 10^{-9} Torr by ion pumping. In order to determine the intensities of the spectral lines in photons per incident ion, it was necessary to calibrate the detection system. A lamp calibrated by the Eppley Laboratories according to the method of the National Bureau of Standards[5] was used as the primary standard for the visible spectrum. A Phillips tungsten-filament lamp was used as the secondary standard because of its more convenient size. In order to extend the calibration to include ultraviolet wavelengths (3000–4000 Å), the branching-ratio method was employed.[6,7] Nitrogen gas was introduced into the target chamber and excited by a 25-keV beam of H^+ or He^+ ions. Observations were made of the relative signals from the second positive system of N_2 and the first negative system of N_2^+. Theoretical predictions of the relative intensities in these two spectral

systems were obtained from the work of Thomas *et al.*[8] and Burns *et al.*[9] Hence the relative sensitivity was established and could be normalized to the absolute sensitivity measured at visible wavelengths using the standard lamp.

II. GENERAL SPECTRAL CHARACTERISTICS

For 20–30-keV He^+ on Cu, Nb, Mo, Ag, and W, there were strong emissions of the following He I lines: 5876 Å ($3^3D \rightarrow 2^3P$), 4472 Å ($4^3D \rightarrow 2^3P$), and 3889 Å ($3^3P \rightarrow 2^3S$). In the case of He^+ on W, there were also weak He I emissions at 4026 Å ($5^3D \rightarrow 2^3P$), 4713 Å ($4^3S \rightarrow 2^3P$), 4922 Å ($4^1D \rightarrow 2^1P$), and 6678 Å ($3^1D \rightarrow 2^1P$). For 20–30-keV H^+ on these same metals, there were emissions of the first three lines of the Balmer series.

For both He^+ and H^+ incidence on Al, we observe a very intense broad band extending from 2500 to 6000 Å, which obliterates the usual lines from scattered projectiles except for He 5876 Å and H_α 6563 Å, respectively. For all other projectile-target combinations, there were various weak broad-band emissions; these will be reported and discussed elsewhere.

Detailed study of the He I lines and H lines indicates that they are of the order of 20–30 Å and 50–60 Å in breadth, respectively, with a sharp peak on the blue side. Figure 1 shows a specimen line shape of a Balmer α transition. The greater breadth of the H lines, compared with He emissions, reflects the greater incident-ion velocity for a given incident energy and therefore a greater Doppler broadening.

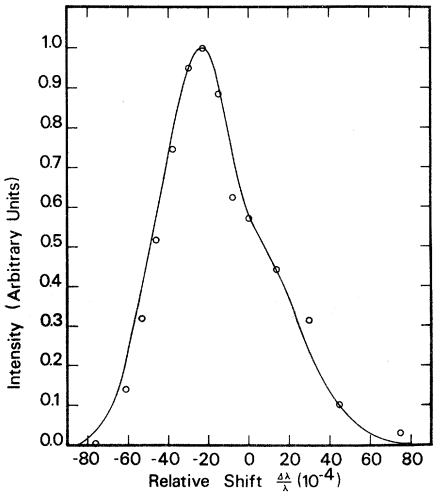

FIG. 1. Measured and predicted line shape of the 6563-Å ($n=3$ to $n=2$) H emission induced by 25-keV H^+ impact on Mo at an incidence angle ϕ of 60°. Intensity is shown as a function of relative wavelength shift, defined as the shift ($\Delta\lambda$) from the 6563-Å line divided by the wavelength of that line, 6563 Å (λ). Circles, experimental data points; solid line, prediction by our model with survival coefficient chosen for best fit to data points ($A/a=7.2 \times 10^7$ cm/sec).

In the case of the aluminum target bombarded by He^+, we observe emissions of Al I lines including all components of the multiplets at 3089 and 3956 Å. For these lines we observe no broadening or wavelength shifts that can be attributed to the Doppler effect. It is therefore concluded that the sputtered excited aluminum atoms are ejected with rather low velocities.

Tests were made to ascertain whether any of these emissions exhibit polarization; this was performed simply by monitoring the light signal as a polaroid analyzer was rotated in front of the monochromator. No evidence of polarization was found.

III. ANALYSIS OF LINE SHAPE

The method for predicting line shape has been described fully in our previous publication[2] and only brief details will be given here. A theoretical prediction by McCracken and Freeman[3] is used to predict the flux and energy distribution of all projectiles emerging from the surface into a particular direction. It assumes that the projectile penetrates into the target, losing energy by electronic stopping and suffering no appreciable deviation; at some point, it suffers a large-angle deviation by a Rutherford-scattering event with a single target atom, and is able to return to the surface. Based on this picture one may formulate an expression for the probability $P(E_S)$ that a projectile will emerge with an energy between E_S and $E_S + dE_S$ and moving into some element of solid angle $d\omega$:

$$P(E_S) = N(E_S)\, dE_S\, d\omega \ . \tag{2}$$

Without any information on the proportion of the scattered projectiles which might be neutralized into a specific excited state, we make the assumption that this proportion, F, is independent of the emergent particle's energy and direction. As we shall see, some of our work tends to support this assumption. The final factor we must account for is the probability that an excited particle will escape from the influence of the surface without undergoing radiationless decay. This is given by Eq. (1). The excited particles that escape will eventually decay radiatively, and a certain fraction F', dependent only on apparatus geometry, will be detected.

Thus, combining these various terms, we have for the probability $P(E_S)$ of detecting a photon from an emergent particle of energy E_S, scattered into

a solid angle $d\omega$,

$$P(E_s) = F' F e^{-A/aV_\perp} N(E_S) dE_S d\omega \ . \quad (3)$$

The wavelength of the photon can be simply calculated by the Doppler-shift formula; Eq. (3) permits a calculation of the relative line shape. In practice, the survival coefficient A/a is unknown; we perform the line-shape calculation for various trial values, and a best fit to experimental data is achieved; from this we establish a "measured" value of A/a.

We have made some studies of how the predicted line shape would be altered if we were to replace the Rutherford-scattering cross section in the calculation[2] by a screened-Coulomb cross section formulated in the manner of Everhart *et al.*[10] This change tends to provide a slightly less satisfactory fit of our predicted curve to the experimental data and does not appreciably change the derived value of the survival coefficient.

IV. RESULTS OF THE LINE-SHAPE ANALYSIS

The consequence of this analysis is that we may determine, from the line shape, a measure of the survival coefficient A/a defined in Eq. (1). We consider that the accuracy with which A/a may be established is poor for incidence angles below 45° due to the low signal strengths and the insensitivity of line shape to the value of A/a adopted; thus, the results quoted here are derived for incidence angles of $\phi \geq 45°$. It has been shown that the derived values are consistent with the experimental data for lower angles of incidence. For large angles ($\phi \geq 45°$) we believe that the survival coefficient can be determined to an accuracy of ±50%; varying the survival coefficient by this amount from the "best-fit" value can shift the wavelength at which maximum intensity occurs by 4 Å or more and so distorts the curve that it bears little resemblance to the measured line shape.

The survival coefficient, A/a, for the $3\,^3D$ state of helium as determined from the 5876-Å He I emission for the cases of Cu, Nb, and Mo targets, was found to be, respectively, 3.0×10^8, 1.3×10^8, and 1.0×10^8 cm/sec. The survival coefficients for the $4\,^3D$ state of helium as determined from the 4472-Å line shape for the cases of Cu and Nb targets were, respectively, 1.6×10^8 and 0.8×10^8 cm/sec. Finally, the survival coefficient for the $3\,^3P$ state, derived from the 3889-Å line shape in the single case of a niobium target, was found to be 1.5×10^8 cm/sec. In all cases these values are the means of several determinations at different energies and impact angles. The above values supersede[11] the measurements we published earlier[2] for some of these cases.

The determinations of survival coefficient for the $n=3$ state of hydrogen, derived from analysis of the Balmer α line shape, yield a mean value of 7.8×10^7 cm/sec for the three metals considered, copper, niobium, and molybdenum. The observed variation between metals is less than the statistical reliability of the data.

In addition to the data listed above we have also studied the line shapes induced by H^+ and He^+ incident on iron and type-304 stainless steel. The survival coefficient A/a for the $n=3$ state of hydrogen is found to be 6.1×10^7 cm/sec; this is less than the value for the other metals studied, but only by an amount which is less than our estimated limits of accuracy. The survival coefficient for the $3\,^3D$ state of helium at a type-304 stainless steel surface is found to be 3.4×10^7 cm/sec; this is significantly less than the values for the other metals studied.

Finally, we would note that we have studied the line shape of the Balmer α line induced by H_2^+ impact on various targets, and find it to be exactly the same as the corresponding situation for H^+ impact at the same velocity. This suggests that the H_2^+ ion dissociates at impact on the surface, and the fragments are uncorrelated in their subsequent behavior.

There have been some limited attempts to calculate theoretically the survival coefficient A/a; these are summarized in convenient form by Hagstrum.[4] Cobas and Lamb[12] calculate separate values for the coefficients A and a for the case of metastable helium atoms being deexcited at a tungsten surface. The calculations employ hydrogenic wave functions and are not expected to be accurate to better than an order of magnitude. The work of Cobas and Lamb,[12] as interpreted by Hagstrum,[4] predicts a survival coefficient of 1.3×10^8 cm/sec for an Auger deexcitation mechanism; this is quite comparable with all the measurements we present here. By contrast, for a process of resonance ionization, the survival coefficient, A/a, predicted[4] by Cobas and Lamb[12] is 4.8×10^{10} cm/sec, and that by Shekhter[13] is 1.8×10^{11} cm/sec. The predictions for a resonant ionization process are three orders of magnitude higher than our measurements.

V. TOTAL INTENSITY OF THE LINE EMISSION

A major objective of this work was to establish an absolute measurement of the total light intensity emitted in a transition; clearly, this can be related to the probability that an incident ion will recoil in a specific excited state. A convenient representation, suggested by Kerkdijk and Thomas,[1] is to define a coefficient γ_{ij} for emission of

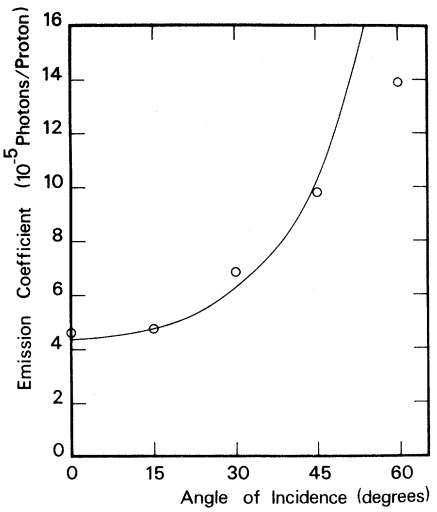

FIG. 2. Emission coefficient of the hydrogen $n=3$ to $n=2$ transition shown as a function of angle of incidence for 25-keV protons incident on Nb. Circles, experimental data points; solid line, predicted dependence with $A/a=7.8\times10^7$ cm/sec. The theoretical curve has been normalized to the data at 15° incidence angle.

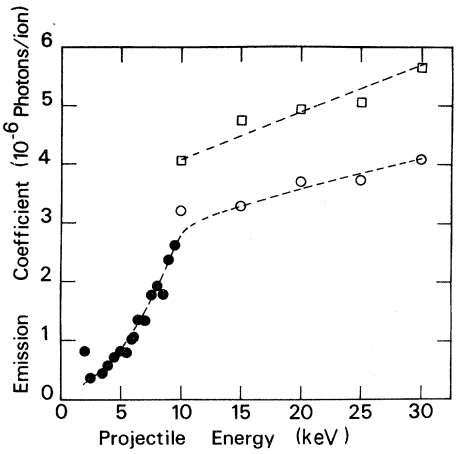

FIG. 3. Emission coefficient γ_{ij} of the helium $3\,^3P \rightarrow 2\,^3S$ transition shown as a function of projectile energy for He^+ ions at an incidence angle of 45°. Square data points for the case of an Mo target, circular points are for Cu. Data below 10 keV are from a previous publication (Ref. 1). The dashed lines are drawn to indicate the general trend of the experimental data.

photons in a transition from state i to state j; this coefficient is simply the total number of photons emitted into all directions per incident projectile. A second valuable factor[1] is the coefficient γ_i for formation of a specific excited state i; this can be defined as the number of backscattered atoms in the state i per incident projectile. In a special case where an excited state i cannot be populated by cascade from higher levels and where the transition $i\rightarrow j$ is the only decay path, then γ_i will equal γ_{ij}. In general this is not true; the state i is populated by cascade from higher levels, and is depopulated by more than one radiative decay path; consequently, γ_i and γ_{ij} are not equal. The general relationship between γ_i and γ_{ij} has been discussed by Kerkdijk and Thomas; moreover, it is analogous to the relationship between excitation and emission cross sections in atomic collision studies.[14]

The emission coefficient γ_{ij} has been measured in this experiment. The line shape of a transition is scanned and integrated; the calibrated sensitivity of the detection system permits this to be converted to an absolute photon flux. We then assume that the emission is isotropic and arrive at the total emission rate by multiplying the photon flux into the spectrometer by $4\pi/\Delta\omega$; here $\Delta\omega$ is the solid angle subtended at the target by the monochromator entrance slit. Taking this photon emission rate and dividing by projectile ion flux, we obtain a value for γ_{ij}.

We estimate a reliability of better than ±10% for the functional dependence on energy and angle of an emission coefficient for a given transition; the principal source of possible error is the statistical variation of signals. In comparing emission coefficients for two different transitions there is an additional source of possible error in the calibration of detection sensitivity at different wavelengths. We estimate the reliability of a ratio between emission coefficients for different transitions to be ±20%; this includes the statistical errors of the individual measurements, and possible errors in the relative calibration of detection sensitivity at the relevant wavelengths. We estimate that the overall reliability of the absolute numerical values is no better than ±50%. This rather large uncertainty arises principally in the absolute calibration of photon-detection efficiency; it includes possible errors in setting the temperature of the primary-standard lamp, and also possible inadequacies in measurement of the relevant geometrical parameters.

In Figs. 2–7 we display some of these measured emission coefficients. They may be shown as a function of incidence angle on the target for a fixed projectile energy (Fig. 2); as a function of impact energy for a fixed incidence angle (Figs. 3–5); or as a function of the target number at a fixed projectile energy and impact angle (Figs. 6 and 7). For measurements involving He^+ on a copper target (Figs. 3 and 4) we show also the earlier relative measurements by Kerkdijk and Thomas; these

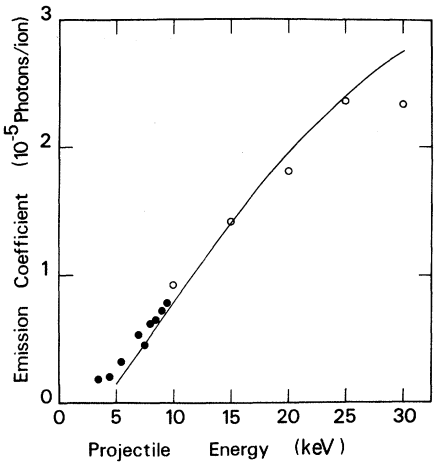

FIG. 4. Emission coefficient γ_{ij} of the helium $3\,^3D \rightarrow 2\,^3P$ transition shown as a function of projectile energy for He^+ ions incident on Cu at an angle of 45°. Data points below 10 keV are from a previous publication (Ref. 1). The solid line is a theoretical prediction normalized to the experimental data at 15 keV and calculated using a survival coefficient, A/a, of 3×10^8 cm/sec.

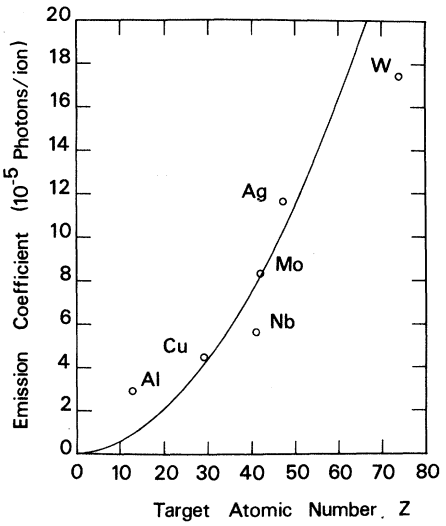

FIG. 6. Emission coefficient of the helium $3\,^3D \rightarrow 2\,^3P$ transition shown as a function of target atomic number. Circles, experimental data points; solid line, McCracken-Freeman model. Experimental conditions: 25-keV He^+ ions incident at 60°. The theoretical curve has been normalized to the data at the Mo data point.

are normalized to the present data at 10-keV impact energy. In certain figures we have drawn lines between the data points to indicate the general trend of the emission coefficient data; such lines, which are specifically identified in the captions, are not meant to imply any detailed knowledge of behavior between the data points.

For comparison purposes we show in Table I the emission coefficients for certain transitions induced by 25-keV H^+ and He^+ on Mo at an angle

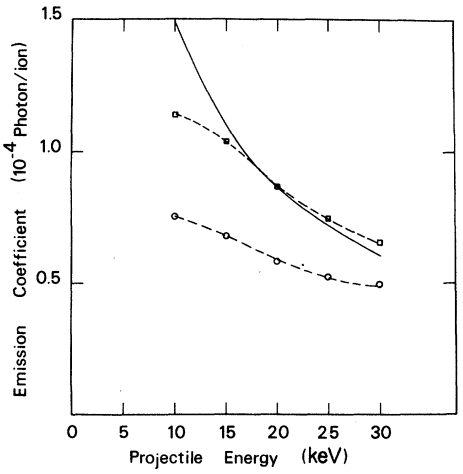

FIG. 5. Emission coefficient of the hydrogen $n=3$ to $n=2$ transition shown as a function of primary energy of H^+ ions at an incidence angle of 45°. Squares are experimental data points for the case of an Mo target; circles, experimental data points for a Cu target; solid line is the predicted dependence for Mo with $A/a=7.8\times10^7$ cm/sec. The theoretical curve has been normalized to the Mo data at 20-keV energy. Dashed lines indicate the general trend of the experimental data.

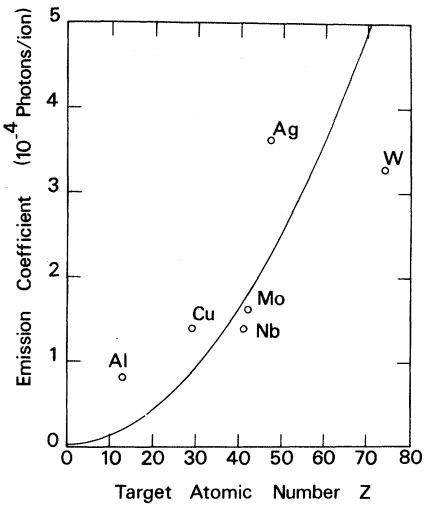

FIG. 7. Emission coefficient of the hydrogen $n=3$ to $n=2$ transition shown as a function of target atomic number. Circles, experimental data points; solid line, McCracken-Freeman model. Experimental conditions: 25-keV H^+ ions incident at 60°. The theoretical curve has been normalized to the data at a single point to provide a best fit.

TABLE I. Measured values of emission coefficient γ_{ij} with estimates of the excitation coefficient γ_i. Data for H^+ and He^+ ions at 25 keV incident on a Mo target at angle of 60° to the surface normal.

Projectile	States i	States j	γ_{ij} (Photons/ion)	γ_i (Excited atoms/ion)
H^+	$n=3$	$n=2$	1.62×10^{-4}	
H^+	$n=4$	$n=2$	1.89×10^{-5}	
H^+	$n=5$	$n=2$	4.92×10^{-6}	
He^+	$3\,^3D$	$2\,^3P$	8.34×10^{-5}	8.17×10^{-5}
He^+	$4\,^3D$	$2\,^3P$	9.04×10^{-6}	1.15×10^{-5}
He^+	$3\,^3P$	$2\,^3S$	9.38×10^{-6}	6.10×10^{-6}

of 60° to the surface normal. In a few cases we have estimated the excitation coefficient γ_i. The excitation coefficient for the $3\,^3P$ state was estimated by allowing for cascade from higher $n\,^3S$ and $n\,^3D$ levels (about 42%), and branching of the decay between $3\,^3P \rightarrow 2\,^3S$ and $3\,^3P \rightarrow 3\,^3S$ transitions; transition probabilities were obtained from the work of Wiese *et al.*[15] For estimates of γ_i for the $4\,^3D$ state, we had no way of assessing cascade and, for the $3\,^3D$ state, could estimate cascade only from the $4\,^3P$ level (about 2%); consequently, the values of γ_i are not really corrected for cascade and include only the branching in the decay transition. For the hydrogen emissions we did not attempt to estimate γ_i; the measured Balmer emission is a sum of three transitions from the almost-degenerate *ns*, *np*, *nd* states to the lower 2*s* and 2*p* levels; there is no way of estimating the relative importance of the separate transitions.

The measured emission coefficients displayed in Table I and in the various figures show that only a small fraction of the projectiles produce photon emission. Most excited atoms formed do in fact decay by nonradiative mechanisms so that the emission coefficient represents only a small proportion of the atoms which were originally excited. It is of interest to estimate what fraction of the backscattered atoms were excited at the point of emergence from the surface; this can only be calculated very roughly. Let us take as an example the case of 25-keV H^+ incident on molybdenum at 60° to the surface normal. Summing the emission function of the first three Balmer lines (Table I), we find the total emission of photons to be 1.9×10^{-4} per incident ion. Using a survival coefficient A/a of 7.8×10^7 cm/sec (the measured value for the $n=3$ state), we estimate that only 21% of all excited atoms in the $n=3$, 4, and 5 levels do in fact radiate, and the remaining 79% decay by the nonradiative mechanism; thus, the total flux of excited atoms formed in these states is about 8.9×10^{-4} per incident ion. Now, it is clear that this figure does not represent all excited atoms, since we do not detect formation of the $n=2$ levels. The work of Sterk *et al.*[16] suggests that formation of $n=2$ levels occurs with about the same likelihood as formation of all the higher excited levels combined. Thus, the formation of excited recoil atoms occurs for only a few projectiles in every 10^3 that are incident on the surface. The conclusion that is to be drawn from these very rough estimates is that less than 1% of all incident projectiles recoil as excited atoms. Inevitably, this means that most backscattered atoms in this case are either ground-state neutrals or ions. A similar conclusion can be drawn from analysis of the case of He^+ impact.

Later in this paper (Sec. VI) we make predictions of the excitation coefficient and compare it with the experimental data; to perform the comparison we make the assumption that the excitation coefficient is proportional to the emission coefficient. The assumption is, of course, exactly true if there is no cascade into the emitting state of interest. The assumption will also be true if the cascading transition, $k \rightarrow i$, exhibits the same functional dependence on impact energy and angle as does the measured transition $i \rightarrow j$. Insofar as we are able to estimate cascade, we conclude that in all cases it has the same functional dependence on energy and angle as the levels which it populates. We shall therefore assume that the excitation coefficients are proportional to emission coefficients.

There are no published data with which these absolute values may be compared; our earlier work gave only the relative variation of emission coefficient with impact energy[1] and angle.[1,2] Shown in Figs. 3 and 4 are earlier measurements of the emission coefficient for HeI transitions induced by He^+ impact on Cu; this relative data[1] has been normalized to the present measurements and appears to join them quite smoothly. We would also note the work of Sterk *et al.*,[16] which measured absolutely the emission coefficient for the Lyman α transition induced by H^+ impact on aluminum. The coefficient was found to be of the order 10^{-3} photons per proton, with an energy dependence generally similar to that displayed in Fig. 5.

VI. THE PREDICTION OF EXCITATION COEFFICIENTS

Equation (3) was used to predict the spectral line shape, and by integration of this shape over all wavelengths one can in fact predict the total intensity of the emission. This can be repeated for various impact angles and energies to arrive at

the functional dependence of the excitation coefficient on these two parameters. Two important parameters are unknown in Eq. (3). We have no theoretical knowledge of the survival coefficient A/a; however, we have our experimentally obtained values listed previously, and we can utilize these for the prediction. Secondly, we have no information on the probability that a backscattered atom will be excited; this is the factor F in Eq. (3). For the purpose of this calculation, we continue to make the assumption that this factor is independent of the emergent particle's energy and direction; success of the predictions would tend to confirm the validity of the assumption. We make no assumption as to the magnitude of this factor, and so our predictions will indicate relative variations only; we therefore normalize theory to experiment in order to perform a comparison.

We do again note that the measured quantity is an emission coefficient, γ_{ij}, and the predicted quantity is an excitation coefficient; our discussion in the preceding section suggests that these two quantities are proportional to each other. Thus, a comparison of the measured γ_{ij} with the predicted γ_i is valid.

In Fig. 2 we show a predicted angular dependence normalized to the experimental data; the agreement between prediction and experiment is very satisfactory. In Figs. 4 and 5 we show the experimental measurements of emission coefficient compared with the theoretical prediction of excitation coefficient. The agreement between prediction and experiment is really quite good, particularly for He^+ on copper (Fig. 4), where good agreement is observed down to 5-keV impact energy.

The agreement between predicted and observed energy dependence in Figs. 4 and 5 is surprisingly good in view of the simplifications made in the prediction. The comparison tends to confirm the assumption that the probability of a scattered particle being formed in a specific excited state is indeed independent of the original incoming projectile's energy. This is consistent with a study by Berkner *et al.*[17] of the excited-state fraction in hydrogen beams that had traversed thin metallic foils; they also conclude that the probability of excited-state formation varies very little with projectile energy.

The energy dependence of the emission coefficient is predicted very well by our model, which involves use of a survival coefficient A/a derived from the line-shape analysis. The predicted energy dependence is a rather sensitive function of the survival coefficient adopted. Consequently, an alternative method of measuring the survival coefficient is to analyze the energy dependence of the emission coefficient. Such a procedure has already been used for the study of excited sputtered particles in the work of White and Tolk.[18]

VII. DEPENDENCE OF EMISSION ON TARGET

A further result of our work is a measure of how the emission coefficient varies with target atomic number. The data are shown in Figs. 6 and 7 for fixed impact energy and angle, for He^+ and H^+ projectiles, respectively. In general, the emission coefficient rises with atomic number although there is some irregularity in the behavior.

It is possible to make a theoretical estimate of this atomic-number dependence, again using the work of McCracken and Freeman.[3] The probability of backscattering [Eq. (2)] with an energy E_S is related to the energy loss suffered by the projectile as it penetrates the target; this energy loss is calculated using the Lindhard-Scharff[19] theory for electronic stopping. The backscattering probability is also related to a Rutherford-scattering event involving the projectile and target nuclei; this large-angle scattering event returns the projectile to the surface. The work of McCracken and Freeman[3] shows that the backscattering probability is proportional to a function $f(Z)$, given by

$$f(Z) = Z_1^{5/6} Z_2 (Z_1^{2/3} + Z_2^{2/3})^{3/2} . \quad (4)$$

Here, Z_1 is the atomic number of the incident ion and Z_2 is the atomic number of the target atom. In effect, the factor $N(E_S)$ of Eqs. (2) and (3) is proportional to the function $f(Z)$. It follows then that a level-excitation coefficient should be proportional to $f(Z)$; if we again make the assumption, justified in Sec. VI, that excitation coefficients are proportional to emission coefficients, then the factor $f(Z)$ will contain the functional dependence of emission coefficient on target atomic number. We have plotted $f(Z)$ in Figs. 6 and 7, normalized to the experimental data.

The agreement between observed and predicted dependence on target atomic number is not entirely satisfactory, but the general trends are the same. Thus, the backscattered excited-atom flux has the same dependence on target atomic number as is predicted for the total backscattered particle flux. It follows then that the excited-state fraction contained in the backscattered flux is not a sensitive function of the target itself; that is to say, the factor F of Eq. (3) is not greatly dependent on the target atomic number. This is consistent with studies by Berkner *et al.*[17] of the excited-atom fraction in projectile beams that had traversed thin metallic foils; they also show that the frac-

tion of excited atoms in the emergent projectile beam is not a sensitive function of the target.

VIII. EMISSION FROM SPUTTERED ATOMS

For the case of He^+ impact on Al (the lightest element which we have used as a target), we observe spectral lines characteristic of excited sputtered Al. These lines do not appear to be Doppler broadened, indicating rather slow-moving atoms. We have investigated the energy dependence of the emission coefficients for two of these lines, and find them to be a decreasing function of primary energy (Fig. 8). We have also observed Al lines in the case of H^+ impact, but only for large angles of incidence (75°).

The two lines considered in Fig. 8 are the only decay paths of the 4^2S level; adding the two coefficients and subtracting cascade population will give the excitation coefficient of the 4^2S level. Cascade from $n=5$ levels contributes about 22% of the excited-state population; cascade from higher levels is negligible. This gives an estimate for the 4^2S level-excitation coefficient of 1.43×10^{-4} excited Al atoms per incident ion; this is for 25-keV He^+ ion impact at an angle of 45°.

The two lines observed, 3944 Å ($4^2S \rightarrow 3^2P^0$, $J=\frac{1}{2}\rightarrow J=\frac{1}{2}$), and 3962 Å ($4^2S \rightarrow 3^2P^0$, $J=\frac{1}{2}\rightarrow J=\frac{3}{2}$), account for all radiative transitions out of the 4^2S level. According to published transition probabilities,[15] the fraction of all transitions represented by the former line is 33%, and by the latter 67%. Using our experimental intensities we find them to be 35% and 65%, respectively.

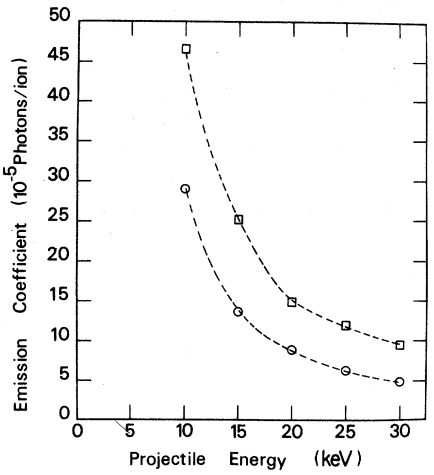

FIG. 8. Emission coefficient of lines from excited sputtered Al as a function of primary energy of He^+ ions (incident at 45°). Squares, experimental data points for Al-3962 Å ($4^2S \rightarrow 3^2P^0$, $J=\frac{1}{2}\rightarrow J=\frac{3}{2}$). Circles, experimental data points for Al-3944 Å ($4^2S \rightarrow 3^2P^0$, $J=\frac{1}{2}\rightarrow J=\frac{1}{2}$). The dashed line is drawn to indicate the general trend of the data.

IX. CONCLUSIONS

The Doppler-broadened spectral lines of H and He observed when H^+ and He^+ ions are incident on metal surfaces have been analyzed by a technique established earlier[2]; the analysis permits measurement of the parameters associated with radiationless deexcitation of excited states at a surface. Using the measured survival coefficients in our simple model, we are able to predict the functional dependence of the level-excitation coefficient on such parameters as projectile impact energy and projectile impact angle. The model also permits a prediction of excitation coefficient as a function of target atomic number. Agreement between experiment and the predictions is very satisfactory in view of the simplicity of the model utilized. We have throughout assumed that the probability of a backscattered atom emerging at the surface in an excited state is independent of its direction, speed, and the nature of the target. This assumption seems to be reasonably valid, and certainly the excited-state population is a very weak function of these parameters. The present experiment also provides measured absolute values of the emission coefficients and some estimates of the absolute values of the level-excitation coefficients. The major gap in our understanding of the whole problem is the absence of any procedure by which the excitation coefficients may be theoretically estimated.

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