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Formation of excited H by impact of 5–30-keV H_1^+, H_2^+, and H_3^+ ions on metal surfaces*

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When 5- to 30-keV H_1^+, H_2^+, and H_3^+ are incident on Mo, Cu, or stainless-steel targets, a small fraction are backscattered in a neutral excited state. These particles subsequently decay and the emission can be observed as Doppler-broadened spectral lines. Measurements of the total Balmer-β line intensity are presented as a function of incoming projectile energy. It is found that H_2^+ and H_3^+ projectiles at energy E behave the same as two or three protons at energy $E/2$ or $E/3$, respectively. The energy dependence of the total intensity down to a projectile energy of 1.67 keV is consistent with a model which includes radiationless deexcitation at the surface.

When light ions such as H^+, with an impact energy between 5 and 30 keV, strike a metal target, a fraction are backscattered in a neutral excited state. The atoms subsequently decay by photon emission which can be observed as Doppler-broadened spectral lines. The line profile depends on the distribution in speed and angle of the scattered particles. Many data have already been collected by Baird *et al.*[1-3] on the profile shape and total intensity of the Balmer-α line emission induced by H^+ impact on a variety of targets (Al, Cu, Nb, Mo, Ag, W). In this work we extend the studies of Baird *et al.*[1-3] by investigating the total intensity of the Balmer-β line emission as a function of incident energy for H^+, H_2^+, and H_3^+ projectiles. The experimental arrangement was identical to that of Baird *et al.*[1] and, therefore, will not be described here.

Theoretical calculations based on a model developed by McCracken and Freeman,[4] and later modified by Baird *et al.*[2,3] successfully predicted the line profile of the emission. The essence of the model is as follows. A projectile penetrating a solid will undergo a single scattering event and return to the surface. Within the target, the particle will lose energy continuously by electronic stopping. As a result of variations in energy loss, penetration depth, and scattering angle, the backscattered flux will exhibit a velocity distribution. However, because the emerging flux contains mostly fast particles ($v > 10^7$ cm/sec), it was suggested[5] that slow particles deexcite near the surface by radiationless decay, presumably, by an Auger deexcitation process.[2,3] For this mechanism, the probability of escaping without radiationless decay is given by

$$P = \exp(-A/av_\perp), \qquad (1)$$

where A/a is a constant defined previously[1-3], and $v_\perp$ is the escape velocity of the particle perpendicular to the surface.

There are two methods of extracting A/a from the data. One can calculate either the observed line shape or the total intensity of the transition as a function of projectile energy. In either case the fitting of theoretical predictions to the experimental data is quite sensitive to the value of A/a adopted, and it is not difficult to choose a suitable A/a which will permit good agreement.

We were able to extend the energy dependence of our data down to 1.67 keV and hence, test the validity of the modified McCracken-Freeman[4] theory in the region 2–10 keV (which was not accessible in the previous work[1-3]). It was shown by Morita *et al.*[6] and Poizat *et al.*[7] that H_2^+ and H_3^+ dissociate on impact, and the fragments are uncorrelated in their subsequent behavior. A similar conclusion was derived by Baird *et al.*,[2] who studied the line shape of the Balmer-α line induced by H_2^+ impact on various targets. They found it to be identical to that induced by H_1^+. Therefore, particles of H_2^+ and H_3^+ with an energy E can be treated as H_1^+ with energy $\frac{1}{2}E$ and $\frac{1}{3}E$, respectively. The relative energy dependence of the experimental data for Cu, Mo, and type-304 stainless-steel targets are shown in Fig. 1. Data for H_2^+ impact at energy E are plotted with the measured intensity divided by 2 at an energy $\frac{1}{2}E$; similarly for H_3^+ the intensity is shown divided by 3 at an energy of $\frac{1}{3}E$. The absence of any systematic difference between data for H^+, H_2^+, and H_3^+ confirms that the molecular projectiles dissociate on impact and the fragments subsequently behave the same as incident H^+ ions of the same velocity. In Fig. 2 the Mo data have been isolated to provide a comparison between experimental and predicted values. Good agreement was obtained with a prediction using Baird's formulation[3] and an A/a value of 1.7×10^8 cm/sec. For convenience, all predictions were normalized to the experimental data at 25

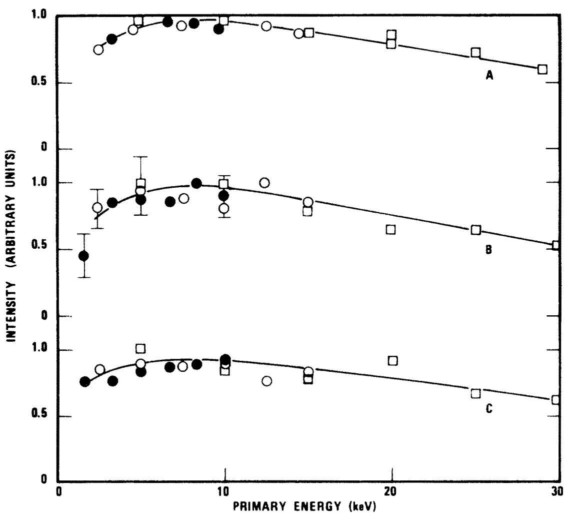

FIG. 1. Energy dependence of Balmer-β line emission from backscattered H* atoms. A, molybdenum target; B, stainless-steel type-304 target; C, copper target. Solid lines indicate the trend of the data. Squares show data for H^+ impact. Open and closed circles are, respectively, data for H_2^+ and H_3^+ impact plotted as described in the text.

keV. In fact, within experimental errors, we found that data for all three targets (Cu, Mo, type-304 stainless steel) could be represented by the computation described by Baird *et al.*[1-3] using A/a values of $(1.7\pm 0.1)\times 10^8$ cm/sec. This suggests that the present theory is adequate down to an incident energy of about 1 keV. Recently, Kerkdijk *et al.*[8] have analyzed the Balmer-β spectral line at impact energies from 10 to 40 keV. They obtained good agreement with experimental data when using an A/a value of $(1.5\pm 0.5)\times 10^8$ cm/sec, which is consistent with our data. Baird *et al.* studied the Balmer-α line and determined an A/a value of 8×10^7 cm/sec from the line shape,[2] and a value of A/a equal to 1×10^8 cm/sec from the energy dependence.[3] The discrepancy between these values is within experimental uncertainties.

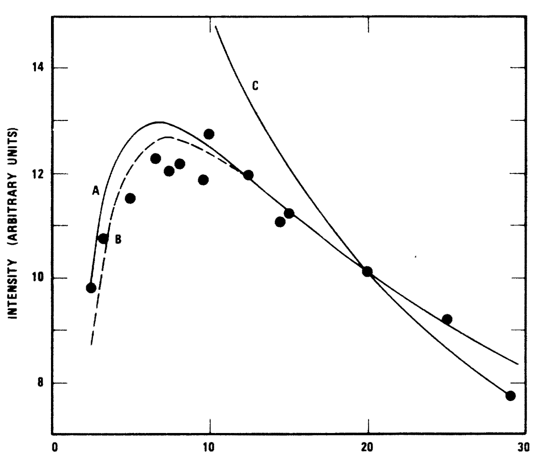

FIG. 2. Experimental and predicted values of the Balmer-β line emission for backscattered H* atoms (Mo target). The circles represent the experimental data. A, theoretical values (without flux attenuation) with $A/a = 1.8\times 10^8$ cm/sec; B, theoretical values (with flux attenuation) with $A/a = 2.1\times 10^8$ cm/sec; C, theoretical values (with flux attenuation) with $A/a = 1.8\times 10^8$ cm/sec.

We also tried to collect data from carbon targets but found the intensity of the Balmer-β emission to be much smaller compared to other targets; the signal-to-noise ratio was close to unity and could not be accurately measured. We estimated that the intensity was consistent with the dependence of the data on atomic number reported by Baird *et al.*[2] In addition we studied the emission from Al_2O_3 targets, but were unable to resolve the Balmer-α or Balmer-β line because of the presence of a very intense broad-band emission.

There are several assumptions upon which the McCracken-Freeman[4] theory is based. Firstly, the probability of a scattered particle being formed in a specific excited state was assumed to be independent of the incident projectile's energy. The good agreement between the predicted and observed energy dependence in the 2-30-keV range seems to confirm this assumption. Secondly, multiple scattering within the target was ignored. So far, no attempt has been made to account for this process. Thirdly, it was assumed that the unscattered projectile flux remained constant with penetration depth. This last assumption can easily be removed. Its effect on the total intensity of the line emission as a function of projectile emission will now be discussed.

The flux of unscattered projectiles should diminish with increasing penetration distance in the solid, because some of the particles are continuously scattered from their original path. We therefore modified the computations of Baird *et al.*[1] to include a flux term which decreased exponentially with distance according to

$$F = F_0 e^{-L/\lambda} ,$$

where L is the penetration depth and $1/\lambda$ is the attenuation coefficient. Although we had no knowledge of the proper attenuation coefficient, we made the reasonable assumption that the unscattered flux F had decreased to 0.01 of its incident value F_0 at a penetration depth equal to the maximum range R; that is, λ was chosen such that $F(L=R)/F_0 = 0.01$. The maximum range R was calculated from Eq. (1) of McCracken and Freeman's work.[4] Figure 2 displays the predicted energy dependence for $F(L=R)/F_0 = 1$ (i.e., no attenuation) and A/a

$= 1.7\times10^8$ cm/sec as well as for $F(L = R)/F_0 = 0.01$ (i.e., with attenuation) at values of $A/a = 1.7\times10^8$ cm/sec and 2.1×10^8 cm/sec. Clearly the prediction without attenuation and $A/a = 1.7\times10^8$ cm/sec and that with attenuation and $A/a = 2.1\times10^8$ cm/sec both represent the data within the limits of experimental accuracy. Thus, introduction of attenuation factors between the two limits defined above can result in the derived value of A/a changing by up to 25%. Analysis of the predictions shows that at an incident projectile energy of 30 keV and with $A/a = 1.7\times10^8$ cm/sec, 90% of the backscattered excited H atoms contributing to the line emissions have an energy of 1 keV or greater when they emerge from the surface. By using Eq. (1) of the McCracken-Freeman[4] theory, it can be shown that these particles (incident on a high-Z target, such as Mo) must have come from a scattering depth no greater than about 2100 Å. For particles with lower primary energies (~1.7 keV), the scattering depth is correspondingly less (~75 Å). In conclusion, we find that because H_2^+ and H_3^+ behave like H_1^+ after impact, we were able to extend our previous data to 1.6 keV. We have shown that all data (H_1^+, H_2^+, H_3^+ on Cu, Mo, and type-304 stainless steel) can be predicted by a modified McCracken-Freeman[4] theory with a survival coefficient A/a of $(1.7\pm0.1)\times10^8$ cm/sec. When a flux attenuation term was included in the theory, it was found to have some influence on the derived A/a coefficient.

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