Higher-Order Z_1 Effects and Effects of Screening by Bound K Electrons on the Electronic Stopping of Channeled Ions*

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The stopping power in {111} Au for channeled ions which have not changed charge has been measured for bare nuclei for $Z = 1$–5 at 3.5 MeV/amu, and for ions bearing 0, 1, and 2 bound electrons for $Z_1 = 1$–9 at 2 MeV/amu. An effective screening of $\sim 0.9e$ per electron is obtained. For the bare nuclei, deviations from simple Z_1^2 scaling are reported. At 2 MeV/amu an increase of 3.5% in S/Z_1^2 is seen from H to He, 3% from He to Li; but no differences are seen from $Z_1 = 5$–9. However, at 3.5-MeV/amu deviations of as much as 20% per Z_1 are observed.

For swift ions totally stripped of electrons in a random medium, the first-order theory of Bethe[1] predicts that the electronic stopping power, S, at a given velocity, v, should be strictly proportional to the square of the ionic charge $Z_1 e$. Observations by Barkas, Dyer, and Heckman[2] on the differences in π^+ and π^- stopping led to the theories of Ashley, Brandt, and Ritchie[3] and Jackson and McCarthy[4] which demonstrated an additive Z_1^3 contribution to the stopping due to polarization. This theory seemed to explain the differences in S/Z_1^2 observed for He and H in various media[5] but Hill and Merzbacher[6] and later Lindhard[7] pointed out that a subtractive Z_1^4 term obtainable from the theory of Bloch[8] must be considered. Hence the expression

$$S = -dE/dx = NZ_2 \frac{4Z_1^2 e^4}{mv^2}(L_0 + Z_1 L_1 + Z_1^2 L_2), \quad (1)$$

where m is the electron mass and N is the density of target atoms Z_2. Lindhard[7] also derived a Z_1^3 contribution somewhat different from that determined earlier[3,4] which, when coupled with the Bloch correction, yielded satisfactory agreement with the He and H results and also fitted the difference between π^+ and π^- stopping. A true test of these contributions requires extension to higher values of Z_1 and accordingly some measurements have been made for Li ions[9] which indicate agreement with Lindhard's approach. However, for higher Z_1 (and to some extent for Li at the energies in question, < 4 MeV/amu), determination of higher-order Z_1 effects is obscured by fluctuations of ionic charge, q, occurring in a random solid. Hence the effect of screening by bound electrons also comes into question.

In this work we utilized our previous observation that an appreciable fraction of O^{8+}, O^{7+}, and O^{6+} ions at ~2 MeV/amu can pass through Au crystal channels with no charge-changing collisions,[10] and that the stopping of these ions in Ag{111} channels is closely proportional to q^2.[11,12] In no case were ions having more than two bound electrons (i.e., L electrons) observed to maintain charge integrity, i.e., O^{5+} exhibited the same stopping as O^{6+} and emerged as O^{5+} by capture at the exit surface (see, e.g., Datz,[13] p. 16). From the electron energy-loss cross sections obtained in Ref. 10 it may be adduced that an electron excited to $n \geq 2$ would be rapidly lost; hence our assumption here that the bound electrons remaining on the ion are in the $n = 1$ state. In this work we use similar techniques but with higher energy resolution, and report the stopping powers in Au{111} channels for totally stripped nuclei of $Z_1 = 1$–9 at 2.0 MeV/amu ($v = 8.95v_0$) and $Z_1 = 1$–5 at 3.5 MeV/amu ($v = 11.8v_0$) (at both energies $Z_1 = 4$ is omitted). The effects of screening were measured at 2 MeV/amu from a comparison of the stopping of totally stripped ions with ions having a single bound electron for $Z_1 = 6$–9 and ions having two bound electrons for $Z_1 = 7$–9.

The ion beams were obtained from the Oak Ridge Tandem Van de Graaff accelerator, stripped, where appropriate, to the desired charge state in a thin gas target, reanalyzed in a second magnet, and passed through a 3715-Å [100] Au single crystal tilted to 40° (path length 4850 Å) and oriented for passage directly through the {111} planar channel. The emergent beam was collimated by a 1-mm aperture 1.5 m downstream and entered an Enge split-pole magnetic spectrograph fitted with a 5-cm-long solid-state position-sensitive detector in the focal plane. Energy calibration was achieved by stepping the beam energy in small increments from the region of anticipated exit energy up to the input energy used in the ex-

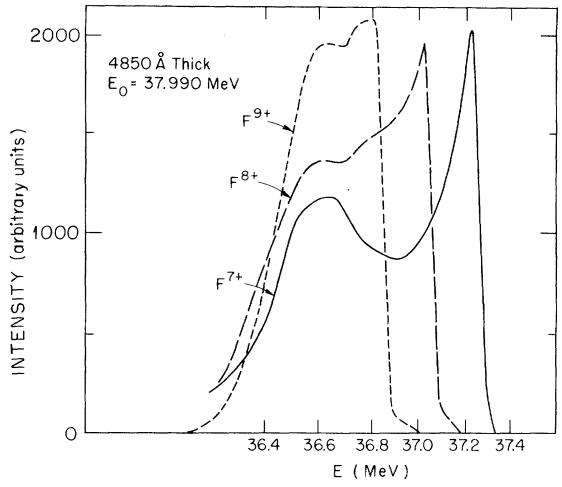

FIG. 1. Energy-loss spectra for F^{q+} ions ($E_0 = 2.0$ MeV/amu) emerging from Au {111} channels. The spectra recorded are for $\Delta q = 0$ (e.g., 9+ in, 9+ out).

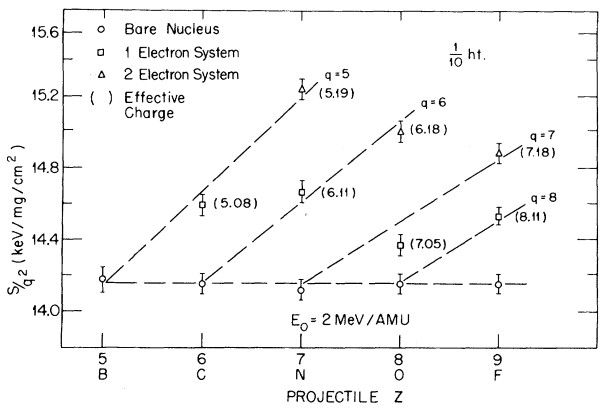

FIG. 2. S/q^2 at $\frac{1}{10}$ height for 2.0-MeV/amu channeled ions with 0, 1, and 2 bound electrons. The dashed lines indicate the connection between ions of the same q.

periment while marking the open beam positions on the detector. The energy resolution of the system was $\Delta E/E = 2\times10^{-4}$ full width at half-maximum. The values of S reported were obtained from the measured $\Delta E/\Delta x$ which was assumed valid at $\frac{1}{2}(E_0+E_1)$ and then back corrected to the initial energy E_0 assuming for channeled ions $S \propto E^{-1}$ in this region. The maximum correction required (that for F^{9+}) was only 1.6%. For random S the curvature appropriate to the region was used.

In all cases reported here the energy-loss spectra were taken for the emergent charge state equal to the input charge state. Such spectra for F^{7+}, F^{8+}, and F^{9+} are shown in Fig. 1. Ions entering parallel to a planar channel follow oscillatory trajectories whose amplitude depends upon the entrance point with respect to the midpoint between planes.[14,15] Ions entering close to a plane (high amplitude) penetrate regions of higher electron density. These particles lose more energy and also equilibrate in charge state.[10] Hence the high energy-loss region is similar for all charge states. The width and the position of the centroid of the peak energy loss can be affected by wavelength restrictions on the emergent particles.[14,15] Particles whose trajectories lie closest to the channel midplanar axis experience the minimum energy loss which contributes to the right-hand leading edge of the spectrum. These are most likely to pass through with no charge exchange. It is these particles which we treat. (To be sure, the energy losses of particles which have even perfectly defined trajectories will not be identical because of statistical straggling, δE. But, insofar as $\delta E \propto \Delta E$, a valid relative comparison of energy losses for center-channeled particles is obtainable from the extrapolated leading edge.) The upper leading edges of all of the spectra in Fig. 1 are extremely sharp down to the ~10% level. The tails observable in the F^{9+} and F^{8+} spectra arise from particles which have captured, then lost an electron along their path. During the time of electron attachment their energy losses are less. The lack of such a tail in the F^{7+} spectrum reflects the improbability of capture into the L shell to form F^{6+}.

The energy loss at $\frac{1}{10}$ height on the leading edge for 0, 1, and 2 electron ions ($Z_1 = 5$–9) divided by q^2 is shown in Fig. 2 for ions at 2 MeV/amu as a function of Z_1. In this region the stopping powers for the bare nuclei are found to scale exactly as Z_1^2 (see below). If the bound electrons screened the nuclear charge perfectly, then, e.g., S/q^2 for B^{5+}, C^{5+}, and N^{5+} should be identical. The higher values of S/q^2 for the clad ions reflects imperfect screening and an "effective" charge $q' = (SZ_1^2/S_z)^{1/2}$ can be obtained by comparison with the stopping power, S_z, of the bare nucleus with the same *charge*. The values of q' are shown in the figure. In all cases studied the screening per electron is ~0.9. Other methods of treating the data, e.g., using the $\frac{1}{2}$ height or extrapolating the leading edge to zero by shape deconvolution will give different values for S/q^2 (see below) but the values of q' obtained are closely the same as those shown in Fig. 2.

Center-channeled ions interact directly only with conduction (6s and 5d) electrons of the Au ($E_b < 10$ eV) and at longer range with 5p, 5s, and

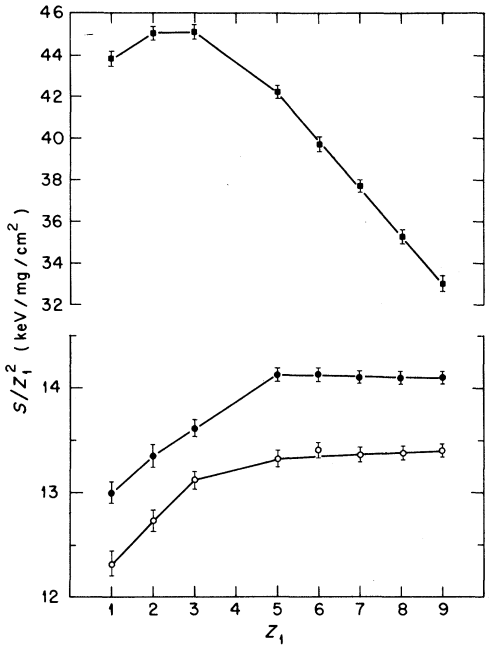

FIG. 3. S/Z_1^2 for 2.0 MeV/amu ions; ● and ○ are leading edge and $\frac{1}{10}$ height values for totally stripped ions in Au {111}, ■ are for random stopping in Au.

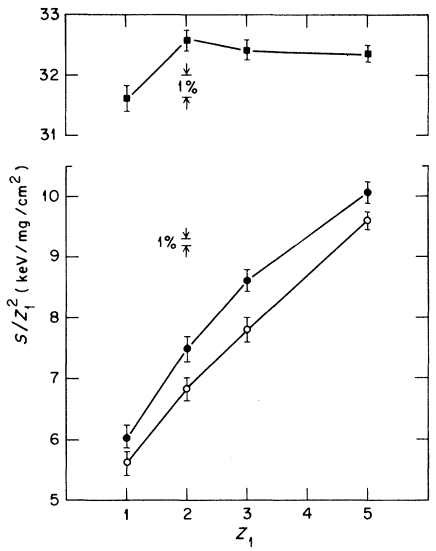

FIG. 4. S/Z_1^2 for 3.5 MeV/amu ions; ● and ○ are leading edge and $\frac{1}{10}$ height values for totally stripped ions in Au {111}, ■ are for random stopping in Au.

$4f$ electrons. The reduction in screening arises from contributions to stopping from conduction electrons which penetrate within the radius of the bound projectile K electron. It might be expected that the effective screening per electron would increase with increasing Z_1 (decreasing orbital radius); however, no such trend can be adduced with the precision of the present data.

In Fig. 3 we have plotted S/Z_1^2 using the extrapolated leading edge and the $\frac{1}{10}$ height of the channeled energy loss spectra for bare nuclei ($Z_1 = 1$–9). For lower values of Z_1, where ΔE is small, a deconvolution of the detector resolution function from the spectrum was necessary in order to obtain the shape of the leading edge. In this we used the general method developed by Anderson.[16] The correction is greatest for the proton data, significant for the He data, and very small for the Li data. The tails on the high-energy portion of the spectra due to ions which have experienced charge-changing collisions (cf. Fig. 1) are present to some extent for all $Z_1 > 5$ and here deconvolution is required to obtain the extrapolated leading edge but not the $\frac{1}{10}$ height. In the same figure we have plotted the peak of the energy-loss spectrum taken in a "random" (nonchanneled) direction in the crystal. The random loss shows a rise (~3%) between H and He expected from theory[3,4,7]; it is flat between He and Li in agreement with Lindhard's prediction,[7] and is in quantitative agreement with the random stopping powers reported by Anderson *et al.*[9] both at this energy and at 3.5 MeV/amu (Fig. 4). At higher Z_1, S/Z_1 declines rapidly because of the increase in the number of electrons which remain bound to the nucleus at equilibrium in random collisions. For the channeled totally stripped ions a rise of 3.5% is seen between H and He, 3.5% between H and Li, only 2% between Li and B, and thereafter no significant change is seen up to F. It is clear that no set of additive corrections of the form given in Eq. (1) can account for this behavior.

At higher velocities (3.5 MeV/amu) the higher-order Z_1 effect is considerably greater (Fig. 4). Here an almost linear increase of almost 20% per unit Z_1 is seen from $Z_1 = 1$–5. The linearity is in accord with a Z_1^3 correction but the magnitude is 6–7 times greater than that for random stopping; this is also in contradiction to the expected decrease in polarization effect with increased velocity. Moreover, if it were to be argued that our assumption of a frozen charge was invalid the probability of bound electrons is larger for larger Z_1 and the real increase in S/Z_1^2 would be even larger.

Only systematic errros are indicated by the error bars; uncertainties due to deconvolution are not included. These are difficult to assess but are only serious for H^+ at 2 MeV/amu and H^+

and He^{++} at 3.5 MeV/amu. In any case, the magnitude of the effect is unassailable since even the undeconvoluted edge of the B^{5+} spectrum at 3.5 MeV/amu lies well to the high energy-loss side of the undeconvoluted peak of the H^+ spectrum.

No theories of higher-order Z_1 effects have been specifically compounded for the case of channeled ions. Theories developed are for random collisions and the analysis for valence electrons has not yet been completed, but it is difficult to see how polarization and Bloch corrections for valence electrons alone can be responsible for the observed effects. Increasing the ion velocity will increase the contribution of inner shells to the total stopping power.[17,18] To first order this contribution is expected to scale as Z_1^2, but polarization terms for large-impact-parameter collisions of channeled ions with $5p$, $5s$, and $4f$ electrons could be large and may be responsible for the increasing Z_1^3 effect at high velocities. The effects of dynamical screening,[19] which have not been considered in detail thus far, also appear to scale only with Z_1^2; however, since the magnitude of this effect which tend to nullify charge differences is larger at lower velocities, deviations from Z_1^2 scaling may tend to suppress the Z_1^3 term for higher Z_1 at lower velocities.

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Classification of Order-Disorder Transitions in Common Adsorbed Systems: Realization of the Four-State Potts Model*

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Order-disorder transitions on square, triangular, and honeycomb arrays are classified according to whether they can be continuous or must be first order by applying a criterion of Landau. In particular, the transitions to 2×2 arrays on triangular and honeybomb lattices are predicted to be in the universality class of the four-state Potts model. A physical realization of this model, N_2 on Kr-plated graphite, is proposed.

In the literature of physical and chemical adsorption, numerous observations of ordered adsorbate structures are reported. Many of these structures are identified as superlattices in registry with the array of adsorption sites provided by the substrate.[1] In only a few cases, however, has the *transition* from the ordered to disordered state been studied.[2-7] With the recent advances