

# Role of Ion-Surface Interaction at the Entry Surface on the Energy Loss of Highly Charged Slow Ions in Solids

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## Abstract

Evidence is obtained from the data of an earlier measurement that the effect of ion-surface interaction on the stopping power of highly charged slow ions is not at all tiny rather remarkably large, even it supersedes the bulk stopping power. The stopping power due to the surface interactions is directly proportional to the charge state of incident ions.

**Keywords:** Highly Charged Slow Ions, Charge Exchange, Surface Potential, Ion Energy Loss

## 1. Introduction

Energy dissipation of charged particles moving through matter [1] has been of interest since the discovery of charged particles. Long ago Ritchie [2] suggested with the basis of surface plasmon theory that a fast electron moving through a foil would lose its energy to both bulk as well as surface. Nevertheless only recently, our experiment [3] showed that stopping power of swift ions through solids can also distinguish these two contributions clearly. Surface stopping power is determined to be only a two order of magnitude smaller than that of the bulk stopping power of the 3.1 MeV/u vanadium ion beam passing through a carbon foil. The surface stopping power is due to the wake potential [4] originated from the surface plasmon. In the past, Koyama *et al.* [5] revealed existence of surface wakefield. They measured a new line in energy spectra of electrons from Al surface by various ions with 0.98 MeV/u ion energy. The electron line energy was larger than that of convoy electron velocity (equal to the projectile ion velocity). Iitaka [6] explained the shifting of convoy peak to a higher energy by image potential of the incident ion. Image charge interaction energies gained by the incident ions in front of the surface is shown to vary with  $Q^{3/2}$ , where  $Q$  = charge state of the incident ion [7]. However, this energy gain is a tiny part of the total energy loss. Further, Schenkel *et al.* [8] experimentally observed that the charge state dependent energy loss of slow ions in solids was not explained with calculated values using the TRIM code [9].

Nevertheless further progress has been made on the ex-

perimental side. A few years back Srivastava *et al.* [10] experimentally observed the surface enhancement in the stopping power of 1 MeV  $N^+$  beam on highly oriented pyrolytic graphite. Recently Papaléo *et al.* [11] reported direct evidence for a strong dependence of the surface modification as a function of charge state of the incident ions. It implies that energy deposition near the surface varies with charge state. With these important evidences we have taken an attempt to look for the origin of such ion-surface phenomena. Underlying interaction leading to energy loss is simply driven by Coulomb's law and hence the ion charge can play a decisive role. There are direct experimental evidences [8,12] showing that higher charge states of the slow incident ions have higher stopping power in solids. This charge state dependence is explained in terms of charge pre-equilibrium effects. However, it has been experimentally shown that such effects do not exist [13,14]. On the contrary, it has recently been shown that the surface wakefield at the exit surface rather plays an important role on the ions [3]. Such a field is not possible to exist for highly charged slow ions as it originates from surface plasmons at the exit side due to passage of swift ions with velocity greater than the Fermi velocity. In this report, we show that some retarding force still exists due to ion-surface interaction that leads to the surface stopping power at the entry surface.

## 2. Background

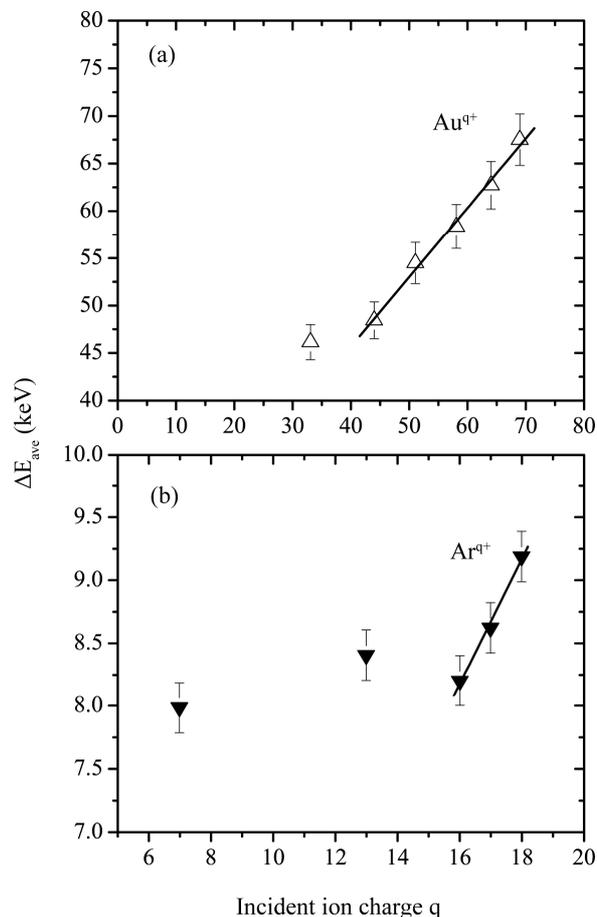
One can plan the energy loss measurements by ions with fixed charge state and constant ion velocity through di-

fferent thicknesses of the foil as done in an earlier experiment [12]. However, it is always a very difficult task to measure foil thickness precisely. Therefore, one can think of an energy loss experiment with different charge states with fixed target foil and constant ion velocity. Further, ions of lower velocity have long interaction time with the Coulomb force. Hence, the highly charged slow ions through very thin foil is the best choice to study the surface field effects on the ions. Such an experiment would be appropriate to know the role of charged states on surface stopping power and was done about a decade ago [8]. We would like to furnish experimental confirmation of the ion-surface effects on energy loss taking results from this experiment [8]. Kinetic energy loss by highly charged slow ions transmitting through thin carbon foils has been measured as a function of projectile charge state from  $q = 3+$  for oxygen to  $q = 69+$  for gold ions. The initial kinetic energies, including relative errors, were  $35.5 (\pm 0.2)$ ,  $92.3 (\pm 0.6)$ ,  $197.7 (\pm 1.0)$ ,  $312.4 (\pm 2)$ , and  $454.4 (\pm 3)$  keV for  $^{16}\text{O}$ ,  $^{40}\text{Ar}$ ,  $^{86}\text{Kr}$ ,  $^{136}\text{Xe}$ , and  $^{197}\text{Au}$  ions, respectively. Ion velocities were low and the same for all the ions ( $v = 0.3v_B$ ) ( $v_B = \text{Bohr velocity}$ ). Several ( $\geq 5$ ) charge states were only used for Ar and Au ions, which have got central attention in this study. Thin carbon foils of  $2 \pm 0.5 \mu\text{g}/\text{cm}^2$  (10.4 nm) were used throughout the experiments.

### 3. Results and Discussions

As mentioned above, no measurements were carried out for the present study rather we made use of the earlier experimental results [8]. **Figure 1** shows the average energy loss values of Ar and Au ions as a function of the projectile charge state,  $q$ . The figure displays clearly that the energy loss values vary considerably with the charge states. However, TRIM code [9] does not take this picture into account, it calculates the energy loss for ions in charge state equilibrium. Calculated energy loss value for gold ions agreed reasonably well with the experimental, average energy loss value for  $\text{Au}^{+33}$  (very far from fully stripped ion). In contrast, the calculated value for Argon ions agreed reasonably well with the average energy loss value for  $\text{Ar}^{+18}$  (fully stripped ion). Thereby, TRIM code is insufficient to represent the energy loss data for slow ions in solid. We thus take an attempt to analyze the data in a different way. One can notice in the figure that the energy loss for  $q = 16, 17,$  and  $18$  for Ar ions and  $q = 44, 51, 58, 64,$  and  $69$  for Au ions show a linear dependence. In contrast, the energy loss for  $q = 7$  and  $13$  for Ar ions (**Figure 1(b)**) and  $q = 33$  for Au ions (**Figure 1(a)**) exhibits different behavior. Energy loss for these ions shoot up from the linear variation.

Different path ways of energy loss are ionization, excitation, and electron capture processes for the present



**Figure 1.** Average energy loss of 2.3 keV/u (a)  $\text{Au}^{33,44,51,58,64,69+}$  and (b)  $\text{Ar}^{7,13,16,17,18+}$  ions in a thin carbon foil (10.4 nm).

ion-target combination as quasi molecular promotion of target electrons to the projectile ions are very unlikely to occur for such asymmetric system [15]. Projectile ionization for high ionic states, that are lying on a straight line in **Figure 1**, is not at all possible as  $v < v_i$ ,  $v =$  projectile ion velocity and  $v_i =$  velocity of the  $i^{\text{th}}$  shell (outer most) electron of the projectile ion. Electron capture cross section ( $\sigma_c$ ) from  $n = 1$  and 2 shells of target atoms to low  $n$  states of these projectile ions is negligible at such a low velocities and is very high to large  $n$  values, as for example,  $\sigma_c = 4.6 \times 10^{-12} \text{ cm}^2$  from  $n = 1$  target shell to  $n = 13$  projectile shell and  $\sigma_c = 2.3 \times 10^{-11} \text{ cm}^2$  from  $n = 2$  target shell to  $n = 26$  projectile shell for  $\text{Au}^{69+}$  [16]. However, such Rydberg electrons cannot survive in the bulk of the foil and thereby no electron capture can occur at all through out the bulk of foil. Therefore, no target ionization is possible by electron capture processes, however, target ionization by direct ionization process will take place equally by all the charge species. The contribution of energy loss from direct ionization can be obtained from

$\Delta E$  intercept of the fitted straight line.

As mentioned already that surface wakefield gave rise to energy loss at the surface [3] and the energy loss due to surface wakefield varies with charge state. Further, we discussed above that nearly neutral atoms emerge out from the exit surface irrespective of the difference in charge state at the incidence. Hence, energy loss dependence with charge states is the outcome of some ion-surface effects at the entry surface. Thus, the ion-surface interaction and direct ionization are the two major mechanisms responsible for the energy loss for slow highly charged ions exhibiting linear charge state dependence.

When the slow ions leave last layer at the exit surface they can be neutralized by capturing electrons at the Rydberg states keeping many inner shells empty. As a result, these hollow neutral atoms will hardly be affected by any electromagnetic interactions at the exit surface. Thus, the observed surface effects must be acting only at the entry surface. Present conclusion is in conformity with an earlier experiment [13] using the  ${}^1H({}^{19}F, \alpha\gamma){}^{16}O$  resonance reaction.

A linear dependence with an intercept as displayed in **Figure 1** represents the contributions coming from two different processes. One comes from a retarding potential due to ion-surface interactions and other from direct ionization. While the former varies with the charge state and the latter does not. For example, the contribution from surface potential and the direct ionization are 49.9 keV and 17.4 keV for  $Au^{69+}$ , respectively. Besides the contribution from the above two mechanisms additional contribution comes from some other process for the charge states showing a departure from linear energy loss processes. This contribution can be obtained by deducting the contribution of the surface wakefield and the direct ionization from the measured value. These values are comparable to the ionization potential of the projectile ions. Therefore, projectile electron loss process is responsible for this. This finding is in agreement with the fact that equilibrium charge states depend on the initial charge states. The equilibrium charge state for these projectile ions can be estimated by equating the sum of ionization potentials with the experimental energy loss contribution due to projectile electron loss process. In case of argon ions the contribution from surface potential and the direct ionization are 7.7 and 1.5 keV for  $A^{18+}$ , respectively. For  $A^{16-18+}$ , no contribution comes from ionization of the projectile ions. However, a large contribution comes from projectile ionization for  $A^{7,13+}$  ions; 4.4 keV for  $A^{7+}$  and 1.5 keV for  $A^{13+}$ . Mean equilibrium charge state within the foil estimated for  $Ar^{+7}$  and  $Ar^{+13}$  are 9.9 and 13.7, respectively. It is worth noting whatever the charge states inside the foil the ions at low velocities will be neutralized at the exit. However, the charge state fraction measurements far from the target

foil will show finite charge states due to multiple Auger-transition cascades. Consequently, the average charge state for  $Au^{69+}$  at an initial velocity of  $0.43 v_B$  was measured about  $1.3 \pm 0.2$  [17].

A remarkable fact is inferred in this work that at low projectile energies, the surface-energy loss is higher than the bulk energy loss. With the increase of projectile energies, the ion will not be neutralized at the exit surface. This fact will result in various charge states of the projectile ion to emerge from the foil, each will then be differently affected by the surface wakefield at the exit. It will cause an uncertainty in the total energy loss leading to energy loss straggling in solid. If the kinetic energy of the incident ion does not reach to the equilibrium charge state at the foil, the energy loss straggling due to the surface wakefield at the exit surface will continue to vary with the foil thickness. Thinner the foil lesser the bulk effect and higher the surface effects, hence, contribution from the surface wakefield to the energy loss straggling shows higher significance for the ultra thin target [18]. Thus, the observed energy straggling is a sum of the statistical fluctuation of energy loss in the bulk and the energy loss distribution of different charge states due to the wake potential at the exit surface. For the relativistic heavy ions, the energy straggling is divided into collisional straggling and charge-exchange straggling. The latter depends critically on the different charge states of out going ions [19]. Since, the charge exchange cross section reduces faster with the ion velocity ( $q^2/v^{15}$  [16]) than the surface-stopping power ( $q/v^2$ ), the energy straggling at surface will play an important role even for the relativistic heavy ions. The contribution from surface is expected to be much higher than that from charge exchange processes.

Energy loss due to the surface potential at the entry surface per unit charge for different ions at  $2.3 \text{ keV}/\mu$  through C-foil are  $0.72 \pm 0.11$ ,  $0.50 \pm 0.14$ , and  $0.06 \pm 0.02$  keV for  $Au^{+69}$ -ions &  $Ar^{+18}$ -ions [8], and protons or antiprotons [20], respectively. Different incident ions give rise to different field strength in the surface potential. Such variation can be attributed to the difference of ion-surface interaction where not only the charge state plays a role but also the ion species take an important role as dielectric properties vary with ion species. Manifestation of such property leads to  $Z_1$  oscillation [21].

We saw that ion-surface interaction results in large surface stopping power for highly charged slow ions. At the entry surface image charge effects and charge exchange processes are the two phenomena known to us. Both are charge state dependent where the former leads to acceleration to the ions and the latter retardation to the ions. Since the net effect is the slowing down, retarding force ought to be larger. Hence, the charge exchange

processes give rise to a surface potential at the entry surface through which ions lose energy as a function of the charge state. This fact is another interesting point in the energy loss of highly charged slow ions in addition to the fact as reported very recently that the unitary-convolution-approximation energy-loss theory explains experimental data well for high to intermediate energies, however, significant deviations occur at low energies [22].

#### 4. Conclusions

We have established that the surface stopping power is directly proportional to the charge state of the ion. Recently, Grüner *et al.* [23] theoretically suggested that the bulk energy loss depends on the charge state due to the charge exchange processes. Interestingly, the charge exchange takes a role in generating a potential as well at the entry surface. The stopping power due to the surface potential varies directly with the charge state. Further, charge state dependent surface energy loss is more prominent than the charge state dependent bulk energy loss for highly charged slow ions. We strongly believe that the energy loss mechanism of highly charged slow ions in solids can be understood better by including a suitable surface potential with the existing theories. The renewed mechanism will help us to understand numerous applications such as, thin-film growth, sputtering, plasma wall interaction in fusion devices, soft-landing, space shuttle glow, detectors, etc.

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