Charge State Dependent Energy Loss of Slow Heavy Ions in Solids

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The loss of kinetic energy of slow ($v = 0.3v_{\mathrm{Bohr}}$) ions transmitted through thin carbon foils has been measured as a function of projectile charge state in the range from $q = 3+$ for oxygen to $q = 69+$ for gold ions. For the first time, charge state dependent energy loss enhancements have been observed for Xe^{q+} and Au^{q+} at $q \geq 40+$, indicating strong preequilibrium contributions to the energy loss of slow, highly charged ions in solids. [S0031-9007(97)04000-3]

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Energetic ions traveling through solids lose kinetic energy in collisions with target electrons and nuclei [1]. Preequilibrium effects and charge state dependencies of energy loss processes of ions in solids have been investigated using beams of light ions [2] and heavy ions at energies ≥ 0.5 MeV/u [3,4], and have been addressed in theoretical studies [5,6]. In this Letter we report on first observations of charge state dependent energy-loss enhancements of slow, highly charged ions in solids.

The interaction of slow, highly charged ions with solids has been studied intensively over the last decade [7–21]. Neutralization of highly charged projectiles above metal surfaces has been described successfully in the framework of a "classical over-the-barrier" model [7]. Having reached a critical distance from a surface, incoming ions begin to resonantly capture electrons into highly excited states, forming "hollow atoms" [10]. Electrons in states with radii in excess of a characteristic surface screening length are "peeled off" [7,8] when the projectile reaches the surface. Quasisimultaneously, a more compact screening cloud is formed around the projectile, and a second, smaller, hollow atom is now formed inside the solid. Currently available estimates of mean deexcitation times, τ_{eq}, of highly charged ions in solids range from less than 0.1 fs (Ar^{16+} in carbon) [16], to a few femtoseconds (bare Ar and Kr ions in aluminum oxide) [10], and 20 fs (Ar^{17+} in carbon) [14]. An upper limit of $\tau_{\mathrm{eq}} \leq 21$ fs has been determined from measurements of equilibrium charge state distributions of slow (2.1 keV/u) Th^{65+} after transmission through $\sim$10 nm thick carbon foils [11].

Reduced screening of the nuclear charge of highly charged projectiles in insulators has been proposed [17,18] to result in increased momentum transfer to target electrons and nuclei. A finite deexcitation time of highly charged ions should result in energy-loss enhancements due to pre-charge-state-equilibrium contributions as compared to the energy loss of projectiles that reach a target in charge state equilibrium.

Density functional theory (DFT) was applied by Juaristi *et al.* [19] to calculate the effect of *K*-shell vacancies on the electronic stopping of slow ions ($Z_1 \leq 16$) in free electron metals. The stopping power for an ion with atomic mass number Z in a medium with given electron density can increase or decrease as a function of *K*-shell vacancies. Oscillations of the stopping power as a function of Z reflect the shell structure of the screening cloud around the ion. The presence of one *K*-shell vacancy, e.g., in oxygen ions is predicted to reduce stopping as compared to oxygen ions with no *K*-shell vacancies by $\sim$40% (in free electron metals with density parameter $r_s = 2$).

Other possible contributions to charge state dependent energy-loss processes stem from the buildup of the screening cloud upon impact of highly charged projectiles on surfaces and from the energy balance in all charge-changing events during deexcitation.

The energy loss of O^{q+} ($q = 2$–7) was found to be nearly independent of the initial ion charge state in small angle scattering experiments using Al(110) single crystal targets [20]. Recently, preliminary evidence for a more pronounced charge state effect was reported for N^{7+} vs N^{6+} [21]. In a study using $Ar^{q+}(q = 8, 12, 16)$ at $v = 0.76v_{\mathrm{Bohr}}$, Herrmann *et al.* found no effect of the initial charge state on projectile energy loss in a 31 nm thick carbon foil [16]. The authors concluded that projectiles reach charge state equilibrium upon penetration of the first monolayer of the target, too fast to enhance the stopping significantly through preequilibrium contributions to energy-loss processes. Up to now, a dependency of the energy loss of slow ions in solids on the initial projectile charge state has not been observed.

We have measured the energy loss of slow ($v = 0.3v_{\mathrm{Bohr}}$), highly charged ions transmitted through thin carbon foils using time-of-flight spectrometry. The setup has previously been described in detail [18,22]. Ions were extracted from the electron beam ion trap (EBIT) at Lawrence Livermore National Laboratory. The pressure in the target chamber was kept below 2.7×10^{-8} Pa. Time-of-flight start signals were provided by secondary electrons emitted from the target at highly charged ion impact. Secondary electrons were detected by an annular

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microchannel plate detector. A negative target bias of -100 V was applied to provide for strong start signals for all initial charge states. The target consisted of a $\sim$10 nm thick ($\sim 2 \pm 0.5$ μg/cm^2) carbon foil [23] with a diameter of 3 mm, mounted on a high transmission grid. The target was tilted at 15°, resulting in an effective thickness, Δx, of $\sim$10.4 nm. The ion flux was typically $<10^3$ ions/s, and the foil was exposed to a dose $<5 \times 10^8$ ions. The energy loss of Xe^{44+} ions was measured at the beginning and at the end of the study, and no indications of foil modification were observed. Transmitted ions were detected by a second microchannel-plate detector after a flight path of 52.5 ($\pm$0.3) cm, and provided time-of-flight stop signals. The solid angle of the detector was 15 msr. Impact parameter selection in transmission experiments results in the preferential suppression of contribution to measured energy-loss values from collisions with small impact parameters in which projectiles are scattered out of the detection angle. At a target thickness of $\sim$50 atomic layers and a collision frequency of $\sim 0.5-1$ collisions per layer, this effect is partially compensated by multiple collisions. Deceleration of positively charged, transmitted ions by the target bias resulted in a small energy reduction in the order of $\sim$1% of the most probable energy loss of ions in the foil. The time resolution of the setup, including detectors and electronics, was about 1 ns. Energy loss in the foil increased ion flight times by $\sim 40-60$ ns, as compared to flight times at the initial ion velocity.

Preparation of projectiles of different charge states and constant impact velocities is critical for this study. Projectile velocities were controlled by acceleration voltages. Standard techniques were used to measure the terminal voltage (6–14 kV) of the EBIT with an uncertainty $\leq 10^{-4}$. In order to limit uncertainties in the effective acceleration voltages due to space charge effects in the ion source [24], we determined projectile velocities additionally by momentum analysis using a 90° bending magnet. A set of apertures was used to collimate the beam after the magnet to a spot size of 1 mm^2. Acceleration potentials were tuned to provide for a constant impact velocity at the target of 6.672×10^5 m/s for gold ions. Probing the ion velocity at the magnet, acceleration by the target bias (-100 V) has not taken effect, and thus the nominal velocities varied from 6.621×10^5 m/s for Au^{69+} to 6.648×10^5 m/s for Au^{33+}. The magnetic rigidity, R, of the analyzing magnet is linearly proportional to the projectile mass-to-charge ratio, m/q. The slope is given by the ion velocity at the magnet: $R \sim v(m/q)$. The value for the velocity resulting from linear regression of the magnetic rigidity data for $Au^{q+}(q = 33, 44, 51, 64, 69+)$ was 6.637×10^5 m/s (standard deviation, 684), consistent with the values of the nominal acceleration voltages. Corresponding measurements were performed for all ion species. A small variation of impact energies stems from the image charge acceleration. The variation is given by $\delta E_{\text{im}} \approx 1.2 \times \Delta q^{1.5}$ (eV) [7,24], and amounts to $\approx$460 eV for Au^{69+} vs Au^{33+}. From these considerations we estimate the overall uncertainty in the impact velocities of projectiles of given mass and different charge states to be $\leq 3 \times 10^{-3}$.

For the investigation of charge state effects on energy loss, we used a velocity of $v = 0.3 v_{\text{Bohr}}$ for all ion species and charge states. The initial kinetic energies, including relative errors, were 35.5 ($\pm$0.2), 92.3 ($\pm$0.6), 197.7 ($\pm$1), 312.4 ($\pm$2), and 454.4 ($\pm$3) keV for ^{16}O-, ^{40}Ar-, ^{86}Kr-, ^{136}Xe-ions, and ^{197}Au-ions, respectively.

Energy-loss distributions of transmitted projectiles are shown in Fig. 1 for Au^{69+} and Au^{33+}. Signal-to-noise ratios in time-of-flight spectra were typically $>3 \times 10^2$. For determination of energy-loss values, measured flight-time distributions, dN/dt, were transformed into energy distributions, dN/dE [25]. Values reported here as average energy losses, ΔE_{ave}, are approximations of the true mean energy-loss values [26,27], and were determined by averaging energy distributions, dN/dE, after subtraction of a constant background. The uncertainty in background determination imposed a limit on contributions from energy-loss events in high-loss tails that could be included in the determination of average energy-loss values. Resulting values of ΔE_{ave} for Au^{33+} and Au^{69+} were 46.0 ($\pm$3) and 67.3 ($\pm$3) keV. Most probable energy-loss values, ΔE_{peak}, were found to be systematically lower than ΔE_{ave} [27] ($\Delta E_{\text{peak}} = 39.1$ and 55.7 keV for Au^{33+} and Au^{69+}, respectively). We note that, due to the relative narrowness of the flight-time distributions, calculations of energy-loss values directly from average flight times yield values that deviate by less than 1% from values calculated after transformation of variables.

Figure 2(a) shows average energy-loss values of oxygen and argon ions as a function of the projectile charge state, q. The error shown in this and the following graph is the relative error resulting from an uncertainty of

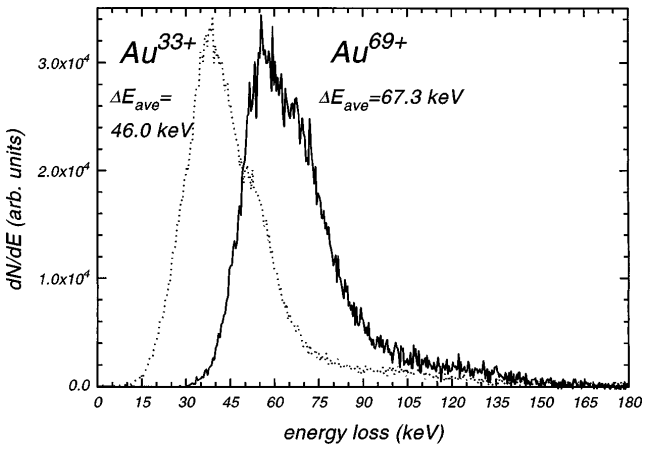

FIG. 1. Energy-loss distributions of Au^{33+} and Au^{69+} after transmission through a thin carbon foil ($\Delta x \approx 10.4$ nm). The initial energy was 454.4 ($\pm$3) keV.

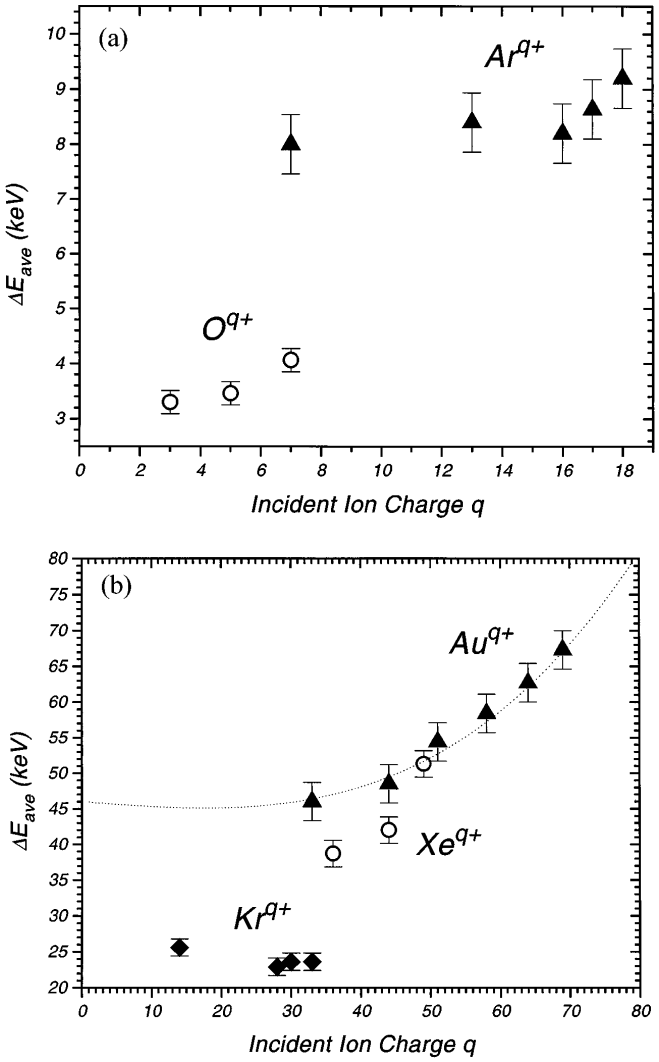

FIG. 2. Average energy loss of (a) $O^{3,5,7+}$ and $Ar^{7,13,16,17,18+}$; (b) $Kr^{14,28,30,33+}$, $Xe^{36,44,49+}$, and $Au^{33,44,51,58,64,69+}$ in a thin carbon foil. The dotted line is a fit to the Au^{q+} data using Eq. (1) with $n = 1$, $m = 2$ ($\alpha = 0.0165$, $\beta = 8 \times 10^{-5}$).

6×10^{-3} in initial ion energy. Average energy losses of O^{7+}, Ar^{17+}, and Ar^{18+} increase slightly over low charge state values. The charge dependent energy-loss enhancements are close to the limit of significance at the given accuracy of the measurements. Using a thinner target, lower ion velocities, and higher ion charge states, our results extend the studies of Ref. [4,16], where no effect of incident ion charge on energy loss was found for F^{8+} and Ar^{16+}, to a regime where the onset of charge effects can be observed.

Predictions from DFT calculations of a decreasing energy loss in the presence of one *K*-shell vacancy in oxygen ions [19] can only tentatively be compared to our results. The calculations considered only electron hole pair excitations of target valence electrons at the Fermi level as mechanism for energy loss. The carbon foils used in this study cannot be described as a free electron gas. Also, our experimental technique does not allow us to differentiate inelastic and elastic contributions to energy-loss processes. Average impact parameters for the later are in the order of the *K*-shell radius of oxygen ions ($\sim$0.14 a.u.). The observed small increase of ΔE_{ave} in the presence of one *K*-shell vacancy in oxygen ions, from 3.5 ($\pm$0.2) keV for O^{5+} to 4.1 ($\pm$0.2) keV for O^{7+} can result from increased momentum transfer in both elastic and inelastic collisions.

Figure 2(b) shows ΔE_{ave} as a function of q for krypton, xenon, and gold ions. For xenon and gold ions, ΔE_{ave} increases strongly with charge. The increase in ΔE_{ave} is 7.6 keV (+33%) as the charge state increases from Xe^{36+} to Xe^{49+}, and 21.3 keV (+46%) as the charge state increases from Au^{33+} to Au^{69+}. This charge state dependent energy-loss increase shows for the first time the presence of strong preequilibrium contributions to the energy loss of slow, heavy ions in conducting solids.

The energy loss in the foil is the sum of contributions to stopping processes before, $(dE/dx)_{\text{pre}}$, and after, $(dE/dx)_{\text{eq}}$, charge state equilibrium is reached. Both equilibration length, Δx_{pre}, and preequilibrium stopping power, $(dE/dx)_{\text{pre}}$, are a function of the projectile charge state, q. In an attempt to estimate charge state dependencies, we assume a simple power law dependency of Δx_{pre} and $(dE/dx)_{\text{pre}}$ on q. This assumption is consistent with results from studies of secondary electron and ion emission from carbon surfaces as a function of projectile charge [11]. We note that a quadratic charge dependency of the electronic stopping power has been assumed in the framework of an effective charge theory in order to estimate charge state effects on the stopping of slow, highly charged ions in solids [16,17]. In a rough approximation that does not include the time dependency of q, we can then write for the energy loss in the foil

$$\Delta E(q) = \left.\frac{dE}{dx}\right|_{\text{eq}} [\Delta x - \Delta x_{\text{pre}}] + \left.\frac{dE}{dx}\right|_{\text{pre}} \cdot \Delta x_{\text{pre}} = \left.\frac{dE}{dx}\right|_{\text{eq}} [\Delta x - \alpha \cdot q^n] + \beta q^{n+m}. \quad (1)$$

The average energy loss value for Au^{33+} agreed reasonably well with values calculated for gold ions in charge state equilibrium using the TRIM code [28]. Thus this value was taken to account for equilibrium energy-loss contributions. With the constants α and β as free parameters, we used Eq. (1) to fit the gold data in Fig. 2(b) for $n, m \leq 2$. The shown fit was obtained for $n = 1$ and $m = 2$. Values for α and β from realistic fits allow for very tentative estimations of Δx_{pre} and $(dE/dx)_{\text{pre}}$. Resulting increases of preequilibrium over equilibrium stopping powers during deexcitation ranged from a factor of 2 along an equilibration length of $\sim$4 nm ($\tau_{\text{eq}} \sim 6$ fs), to a factor of 5 with $\Delta x_{\text{pre}} \sim 1$ nm ($\tau_{\text{eq}} \sim 2$ fs). The observed charge state dependent energy-loss enhancement can be interpreted as resulting from increased momentum transfer to target electrons and nuclei in collisions

with impact parameters comparable to the radii of unoccupied levels in transient hollow atoms. The interaction potential changes on the time scale of the collision time ($\sim$0.5 fs) due to rapid deexcitation of the projectile. In the absence of a critical number of inner shell vacancies, e.g., for ions like Kr^{33+}, deexcitation processes at and below the surface are too fast ($\tau_{eq} \sim 1$ fs, $\Delta x_{pre} \sim 0.7$ nm) and preequilibrium stopping power increases are too weak ($dE/dx|_{pre} < 2dE/dx|_{eq}$) to allow for strong contributions to energy-loss processes before deexcitation is complicated. Contributions to energy loss from the large ($>q$) number of charge-changing events are expected to be significant, but cannot be quantified on the base of our results. Measurements of preequilibrium energy loss contributions in coincidence with detection of characteristic secondary particles (e.g., target and projectile x rays) and as a function of target properties (e.g., atomic mass number and electrical properties) show promise as means to quantitatively differentiate deexcitation channels in the interaction of slow, highly charged ions with solids.

In summary, the average energy loss of slow, highly charged ions in thin carbon foils has been measured as a function of the projectile charge state. For the first time, enhancements of average energy-loss values as a function of ion charge were observed for Xe^{q+} and Au^{q+}, at $q \geq 40+$. The results indicate strong pre-charge-state-equilibrium contributions to energy-loss processes due to the finite deexcitation time of highly charged ions in solids.

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