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Energy loss of slow, highly charged ions in solids

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The loss of kinetic energy of highly charged, heavy ions (Ar^{18+}, Xe^{44+}, and Au^{69+}) in thin carbon foils has been measured as a function projectile velocity in the range from 6×10^5 to 11×10^5 m/s. Evidence for strong pre-equilibrium energy-loss enhancements is observed for highly charged xenon and gold ions. [S1050-2947(97)50809-2]

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Energetic ions traveling in solids lose kinetic energy in collisions with target electrons and nuclei [1]. Energy loss is a function of the projectile charge state in the solid [2]. Pre-charge-state equilibrium effects have been observed in the high-energy regime (>0.5 MeV/u), where projectiles entering solids with charge states lower or higher than the mean equilibrium charge show increased or decreased stopping until charge-state equilibrium is established [2,3]. Stopping power increases in the first few atomic layers of solids have been reported also for N^+ and He^+ ions at relatively low kinetic energies (i.e., 300 keV) [4]. The interpretation of this effect in terms of ''enhanced pre-equilibrium'' stopping has been discussed controversially [5]. Pre-equilibrium effects on the stopping and ranges of slow, highly charged ions were investigated by implantation of Xe^{44+} (0.8–2.3 keV/u, $v=0.18-0.3v_{\mathrm{Bohr}}$) into thermal SiO_2 films on Si [6], but no effect of the ion charge on the range was observed. Analysis of the interaction potentials led to predictions of strong, pre-equilibrium nuclear stopping power increases for the interaction of very slow, highly charged ions with ionic insulators. Evidence for the latter was reported from a study using Ar^{16+} ($v\approx0.3v_{\mathrm{Bohr}}$) transmitted through thin CaF_2 films [7]. No charge effect on stopping was observed in measurements of the energy loss of Ar^{q+} ($q=8,12,16+$) at $v=0.75v_{\mathrm{Bohr}}$ in thin carbon foils, indicating that charge-state equilibrium was established upon penetration of the first monolayer of this semimetallic target and within a deexcitation time of a fraction of 1 fs [8].

Highly charged ions at velocities below the Bohr velocity have charge states very far from mean equilibrium charge states [9]. Relaxation of these ions into charge-state equilibrium requires a finite, currently not well known, deexcitation time [10]. In this article we report on observations of pre-equilibrium effects in measurements of the energy loss of slow, highly charged ions in thin carbon foils. We have studied energy loss as a function of projectile velocity for bare argon and neonlike xenon and gold ions in the velocity range from $6\times10^5-11\times10^5$ m/s. A report on charge-state-dependent energy-loss increases has been given elsewhere [11].

Slow, highly charged ions were extracted from the electron beam ion trap (EBIT) at Lawrence Livermore National Laboratory [12]. Projectiles reached the scattering chamber after momentum analysis in a 90° bending magnet. The ion beam was collimated after the magnet to a diameter of 1 mm. The target consisted of a thin carbon foil with a nominal thickness of 10 nm (2 ± 0.5 μg/cm^2) [13] and was tilted at 15°, resulting in an effective thickness of ~10.4 nm. The total ion dose used in this study was $<5\times10^8$. Repeated energy-loss measurements under identical conditions showed no signs of foil modification in the course of the study. The experimental setup has previously been described in detail [7,12]. Electrons emitted from the target upon impact of individual projectiles were detected by an annular microchannel plate detector (MCP) and provided start signals for the time-of-flight (TOF) measurements. A bias of -100 V was applied to the target to provide for strong start pulses for all projectiles. Transmitted ions are detected after a flight path of 52.5(±0.3) cm by a second MCP and provided time-of-flight stop signals. The solid angle of the detector was 15 msr. Selection of impact parameters in transmission geometry can result in a preferential suppression of the contribution to energy loss from small-impact-parameter collisions, where projectiles are scattered out of the detection angle. At a target thickness of ~50 atomic layers and at an estimated collision frequency of 0.5–1 collisions per atomic layer, this effect is partially compensated by multiple collisions [14]. Impact velocities were controlled by acceleration voltages. Standard techniques were used to measure the EBIT terminal voltages ($U_{\mathrm{terminal}}=3-16$ kV) with a fractional uncertainty of less than 10^{-4}. Acceleration of incident ions by the target bias was included when tuning impact velocities. Deceleration of transmitted, mostly singly positively charged ions by the target bias results in a small energy reduction in the order of $\sim1\%$ of the most probable energy loss in the foil. The uncertainty in initial kinetic energies was estimated by comparison of values from measured acceleration voltages and results from momentum analysis of projectiles using the 90° bending magnet. Including a small variation due to different image charge accelerations [15], the uncertainty in impact energy was 0.6%, or 3 keV, at an impact energy of 454 keV. Energy loss in the foil resulted in flight time increases of 40–60 ns. The time resolution of the setup was ~1 ns.

Energy-loss values were determined from energy-loss distributions, dN/dE, after transformation of variables from measured flight time distributions, dN/dt [16]. Values reported here as average energy-loss values, ΔE_{ave}, are approximations of the true, mean energy-loss values [17] and were obtained after subtraction of a constant background. Signal-to-noise ratios in TOF spectra were typically >300. The uncertainty in background determination limited contri-

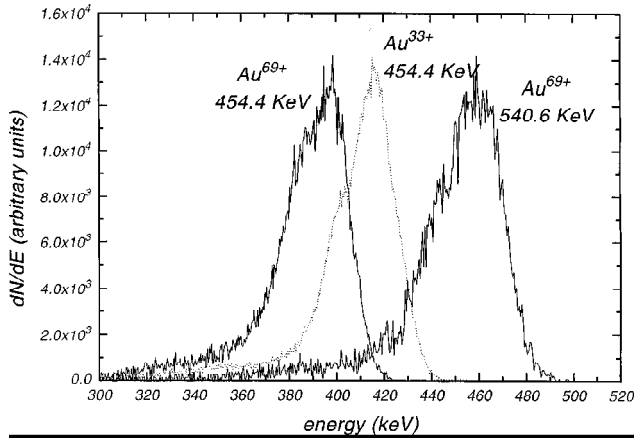

FIG. 1. Energy-loss distributions, dN/dE, of Au^{33+} and Au^{69+} ions transmitted through an $\sim$10.4-nm-thick carbon foil. Impact energies were 454.4($\pm$3 keV) ($v=6.67\times10^5$ m/s) for Au^{33+} and 454.4 and 540.6 ($v=7.3\times10^5$ m/s) for Au^{69+}.

butions in the high loss tails that could be included in the determination of $\Delta E_{\rm ave}$ values. Most probable energy-loss values were found to be systematically lower than $\Delta E_{\rm ave}$. Figure 1 shows dN/dE for Au^{33+} at an impact energy E_0 of 454.4($\pm$3) KeV and for Au^{69+} at $E_0=454.4$ and 540.6 ($\pm$3.2) keV. The energy loss is found to increase strongly as a function of initial projectile charge state [11] and, for Au^{69+}, as a function of impact energy. The charge-dependent increase amounts to 46% (46 keV for Au^{33+} vs 67.3 keV for Au^{69+}). $\Delta E_{\rm ave}$ for Au^{33+} agrees reasonably well (i.e., within the large uncertainty in the value of the foil area density) with energy-loss values estimated using the TRIM code [18] [see Fig. 2(b)]. The dependence of $\Delta E_{\rm ave}$ on the impact velocity v is shown in Fig. 2(a) for Ar^{18+} and Xe^{44+} and in (b) for Au^{69+}. The error bars reflect the relative errors in $\Delta E_{\rm ave}$ resulting from an uncertainty of 0.6% in impact energy. For ions in charge-state equilibrium in this velocity regime, contributions from elastic collisions are significant or dominant, but decrease with increasing impact velocity [18]. Inelastic energy loss of ions in solids has been found to increase proportional to the projectile velocity, $S_{\rm el}\sim v^n$, with $n=0.7$–1 [18–20]. To allow for a comparison with the stopping of ions in charge-state equilibrium, we use values for nuclear, S_n, and electronic stopping powers, $S_{\rm el}$, as calculated by the TRIM code [18]. Resulting values, multiplied by the effective target thickness, are shown in Fig. 2(b) for slow gold ions in carbon. Stopping-power values calculated using TRIM cannot expected to be very accurate in this velocity regime, but have been shown to reproduce the velocity dependencies of elastic and inelastic contributions to energy loss in conducting, monatomic solids correctly. The energy loss of Au^{69+} is found to increase strongly as a function of velocity. The increase is significantly steeper than the increase of $S_{\rm el}$ for ions in equilibrium alone. The sum of contributions from elastic and inelastic energy loss processes increases only very little (i.e., by $<10\%$) in the small velocity range probed in this study. Average energy-loss values for Ar^{18+} are found to be nearly constant when the impact velocity is changed from 6.7×10^5 to 11×10^5 m/s. This can be attributed to a compensation of decreasing nuclear and increasing electronic contributions to energy-loss processes

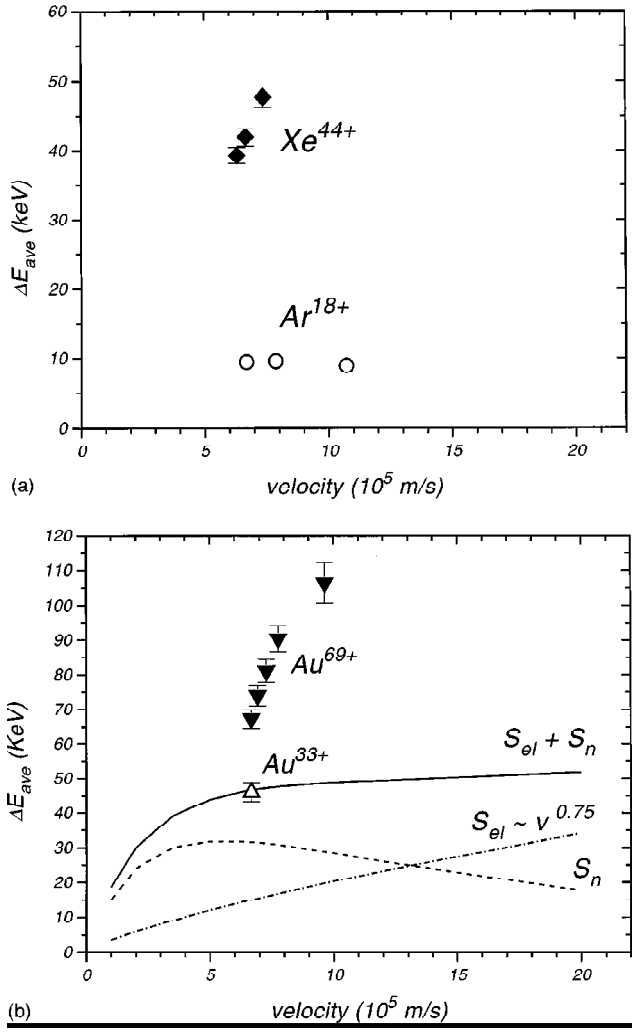

FIG. 2. Average energy loss $\Delta E_{\rm ave}$ of (a) Ar^{18+} and Xe^{44+} and (b) Au^{69+} in a thin carbon foil as a function of projectile velocity. The lines are estimated energy-loss values of gold ions in charge-state equilibrium, calculated by TRIM [18]. ·-·-, S_n, nuclear energy loss; - - -, $S_{\rm el}$, electronic energy loss; —, sum of S_n and $S_{\rm el}$.

[18,20]. $\Delta E_{\rm ave}$ for Xe^{44+} increases also, but the increase is weaker than that for Au^{69+}.

We interpret the strong increase in the stopping of Au^{69+} and Xe^{44+} with impact velocity as resulting from pre-equilibrium energy-loss enhancements. This interpretation is consistent with recent observations of charge-state-dependent energy-loss increases of slow, highly charged ions in solids [11]. Highly charged ions form hollow atoms above metallic and insulating surfaces [21]. At impact on a target, electrons in Rydberg states are peeled off. Quasisimultaneously a screening cloud of target electrons is built up around the projectile and a more compact hollow atom is formed inside the solid [22,23]. Neutralization of the projectile charge by this screening cloud can require extreme degrees of target polarization, involving, e.g., over ten carbon atoms to provide the charge required to neutralize a Au^{69+} projectile. Deexcitation of hollow atoms in solids proceeds via Auger cascades [22] and radiative transitions [21,23]. Available estimates of total deexcitation times $\tau_{\rm eq}$ range from less than 1 fs (Ar^{16+}) [8], to a few femtoseconds (bare argon, iron, and krypton) [21] to an upper limit of 21 fs for

ions up to Th^{65+} in carbon [10]. In the asymmetric combination of a heavy projectile ($Z_{\text{projectile}}=79$) incident on a light target ($Z_{\text{target}}=6$), direct filling of M-shell vacancies in gold projectiles by target electrons is strongly inhibited. Only projectile levels with principal quantum numbers $n\simeq 6-10$ can be populated directly. The charge distribution in the hollow atom is characterized by electrons in the initially filled K and L shells and by electrons in excited states. Intermediate levels with $n=3-5$ start out empty and are filled in the course of deexcitation. Screening of the projectile nuclear charge is reduced at distances from the nucleus equal to the radii of these empty levels. Averaging over all impact parameters, this can be interpreted as an increased effective charge of hollow atoms. Scaling of the electronic stopping power with the square of an effective ion charge was proposed to estimate charge-state-dependent electronic stopping-power increases for slow, highly charged ions [6,8]. At atomic transition rates on the order of $10^{15}-10^{16}$ s^{-1} [21], screening of the nuclear charge and consequently also the interaction potential between projectile and target nuclei change on the time scale of individual collisions (~ 0.5 fs). Momentum transfer to target electrons and nuclei in collisions with impact parameters comparable to the radii of unoccupied levels in the transient hollow atom is increased over corresponding values for projectiles in equilibrium. Increasing the impact velocity of Au^{69+} allows for the deexciting projectile to engage in more collisions before a dynamic charge-state equilibrium is established. The pronounced velocity-dependent energy-loss increase is thus indicative of pre-equilibrium energy-loss enhancements. Increasing the velocity further will extend deexcitation deeper into the target and will eventually allow projectiles to exit the foil highly excited and in mean charge states in excess of equilibrium charge states. The effect is less pronounced for Xe^{44+}, as the pre-equilibrium stopping increase is weaker and the mean deexcitation time is shorter than for Au^{69+}. In agreement with results from studies of the charge-state dependence of the energy loss [11], no indications for strong pre-equilibrium effects are observed for Ar^{18+}. From the charge-state dependence of the energy loss [11], we have estimated the increase of pre-equilibrium over equilibrium stopping powers and the mean deexcitation time for Au^{69+} in carbon. Values ranged from a pre-equilibrium stopping increase of a factor of 5 during a deexcitation time of only 1 fs, to a stopping increase of a factor of 2 over 5 fs. For Ar^{18+} we found an upper limit for the pre-equilibrium stopping increase of a factor of 2 during a deexcitation time of 1 fs. The velocity dependencies of ΔE_{ave} are consistent with our earlier results. The pre-equilibrium effects of ion charge and velocity combined result in energy-loss enhancements for Au^{69+} in thin carbon foils of over a factor of 2. Contributions from inelastic and elastic energy-loss processes as well as the energy balance in the large ($>q$) number of charge changing events cannot be distinguished on the basis of our results.

In summary, the energy loss of slow, highly charged ions in a thin carbon foil has been measured as a function of impact velocity and for different projectile charge states. Both the velocity dependence of the energy loss for Xe^{44+} and Au^{69+} and its increase with ion charge (for Au^{33+} to Au^{69+}) demonstrate the presence of strong pre-equilibrium energy-loss enhancements. Pre-equilibrium contributions to energy loss stem from the finite deexcitation time of highly charged ions in solids.

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