Strong Evidence for Enhanced Multiple Electron Capture from Surfaces in 46 MeV/u Pb^{81+} Collisions with Thin Carbon Foils

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Strong evidence has been found for enhanced multiple electron capture into 46 MeV/u Pb^{81+} with a significant contribution from the entrance surface of thin carbon foils. Capture of up to five electrons has been observed. The multiple electron capture yield is found to increase with decreasing target thickness for thin targets. A simple model describing the data and showing the importance of capture from surfaces is discussed. Further evidence is found for a pronounced asymmetry between electron capture at the entrance and the exit surfaces. Absolute yields for multiple electron capture and projectile ionization are presented. The experimental total cross sections for single capture and ionization agree well with theory.

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The interaction of slow, highly charged ions with solids has attracted increasing interest in recent years, also due to possible future applications. In contrast to pure ion-atom collisions as in gaseous targets, the ion-solid interaction allows one to study the dynamics of atomic collisions in the presence of neighboring atoms. So called "solid-state effects" on the collision are still not fully understood [1–6]. Besides the interaction of the ion with the bulk material, it has been shown that for slow, highly charged ions the interaction with the surface plays an important role (see, e.g., [7] for an overview). Before entering the target, the ion may polarize it and extract electrons from the surface via electron capture into high lying states. Even complete neutralization can be reached and so called "hollow atoms" are formed [8–11]. These surface effects have so far been studied mostly for bare, very slow light ions (typically $Z = 18$) in the keV energy region available from electron-cyclotron-resonance sources. However, with these slow ions only the interaction with the entrance surface could be studied.

The SIS/ESR accelerator and storage ring facility at Gesellschaft für Schwerionenforschung Darmstadt m.b.H., Darmstadt, allows one for the first time to study highly charged (bare, hydrogenlike, etc.) very heavy ions ($Z\alpha \rightarrow 1$, α being the fine structure constant) and their interaction with matter at moderately low velocities. For high $Z\alpha$ ions radiative transitions dominate the decay of excited states in contrast to low and medium heavy ions where primarily nonradiative transitions lead to autoionization. Using the Accel-Decel-Technique [12] beams of hydrogenlike ions up to uranium with energies presently down to below 15 MeV/u and intensities in the order of 10^5 ions/s are available. For the K shell (L shell) of uranium this energy corresponds to an adiabaticity parameter of $\eta_K = 0.06$ ($\eta_L = 0.27$), where η_i is the ratio between the kinetic energy of an electron moving with the projectile velocity and that of the bound electron.

In this paper we present the first measurements on the charge state distribution of hydrogenlike Pb ions at a moderately low energy of 46 MeV/u ($\eta_K = 0.25$ and $\eta_L = 1.03$) after passage through thin carbon foils of varying thickness.

For the experiment, lead ions delivered by SIS with 250 MeV/u were stripped to bare ions and then injected into the ESR storage ring. After deceleration to 46 MeV/u and cooling in the ESR, Pb^{81+} ions were extracted towards the experimental area utilizing charge exchange in the cooler section [12]. Behind the target (see Fig. 1), the beam was refocused in the horizontal plane with a quadrupole doublet and the various charge states were separated in a bending magnet. The ions were detected about 8 m downstream of the target on a two dimensional position sensitive microchannel plate (MCP) detector with fast delay line readout [13] allowing one to measure up to eight charge states simultaneously. With a typical vacuum better than 10^{-6} mbar the equivalent target thickness of the residual gas in the beam line between the last bending magnet in front of the target and the spectrometer is less than 2×10^{-3} μg/cm^2. A test run with an empty target has shown no perceptible background contribution.

Figure 1 shows the charge state distributions for projectiles traversing 11 and 46 μg/cm^2 thick carbon targets.

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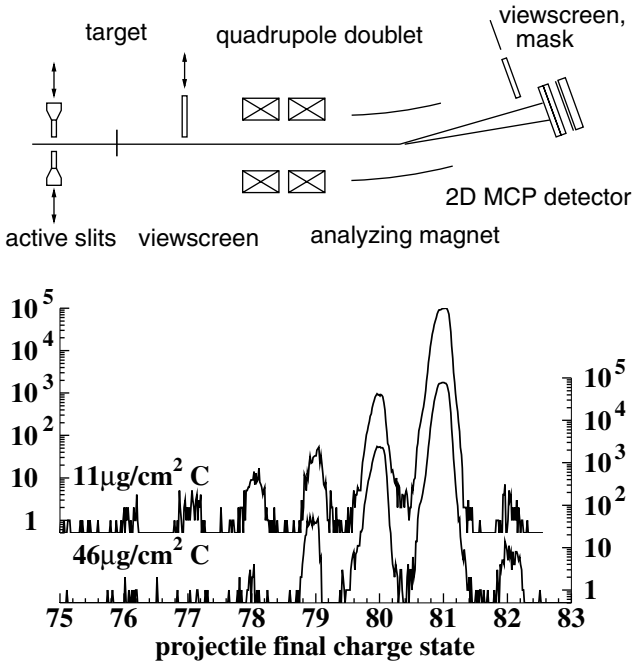

FIG. 1. Experimental setup and charge state distributions as detected on the projectile detector for 46 MeV/u Pb^{81+} on 11 $\mu g/cm^2$ (top) and 46 $\mu g/cm^2$ (bottom) C foils.

The most intense line comes from the direct beam. To the right of it we find bare lead ions resulting from projectile ionization in the target. The multiple electron capture lines on the left side (charge states 76–79) are clearly more prominent in the thinner target.

For thin targets the cross sections for single capture and projectile ionization can be directly determined by $\sigma_q = \frac{N_q}{N_0 \Delta x n}$, where N_q is the number of counts with charge state q, N_0 is the total number of counts in the spectrum (corresponding to the total beam intensity), Δx is the linear target thickness, and n is the particle density. For electron loss the measured cross section of $\sigma_{\text{loss}} = 54 \pm 4$ b is in good agreement with semiclassical approximation calculations using the program IONHYD by Trautman *et al.* [14], which gives a value of 60 b. No dependence of the cross section on the target thickness is found, demonstrating for this process single collision conditions.

The same holds true for the dominant single electron capture, where a cross section of $\sigma_{1-\text{cap}} = 13.3 \pm 0.7$ kb is obtained. Measuring the x-ray emission [15] we find a cross section for radiative electron capture into the K shell (K-REC) of $\sigma_{K\text{-REC}} = 1.6$ kb. The cross section for REC into higher shells is significantly smaller [16]. The remaining cross section for nonradiative capture is in reasonable agreement with calculations in the eikonal approximation [17,18]. In this approximation the cross section has its maximum for capture into the $n = 5$ shell. As we find no dependence of the measured single electron capture cross section on the target thickness—cf. Fig. 2—the reionization probability is not important for the target thicknesses

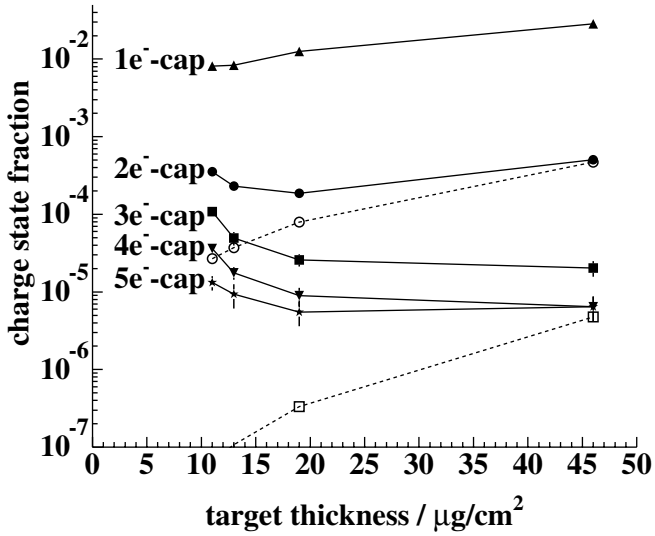

FIG. 2. Measured fraction of ions which have captured one to five electrons as a function of target thickness for 46 MeV/u Pb^{81+} on C. The open symbols give the normalized square and cube of the single capture fraction (see text). The lines are meant to guide the eye only.

used. Based on the experimental error bars the very upper limit for reionization of electrons from single capture is 10 kb which is in the same order of magnitude as the 5s ionization cross section of 8.3 kb calculated with the IONHYD program [14].

In Fig. 2 the measured charge state fractions ($P_q = N_q/N_0$) for capture of up to five electrons are displayed. For double and more electron capture at first a steep drop in the measured fractions with increasing target thickness is observed. Then for double capture the fraction increases again, whereas for triple and more electron capture it seems to level off. This is in strong contrast to what is expected.

True multicapture events in the bulk should lead also to a linear increase of the corresponding charge state fractions with target thickness. Moreover, considering similar collision systems with gaseous targets those events should not dominate [19–21]. Multiple electron capture may not be described by one-step cross sections alone, as it can also be the result of successive single capture events inside the bulk. If successive single capture is the only capture process the probability for detection of n-electron capture is given by $P_n = P_1^n/n!$ with P_1 the single electron capture probability.

In this case double and triple capture should vary, respectively, like the square and the third power of the target thickness—see dotted lines in Fig. 2—which is in obvious contrast to the observation. For instance, for triple capture with the thinnest target $P_1^3/6$ is 3 orders of magnitude smaller than the measured fraction P_3.

In order to understand these unexpected dependences of the multielectron capture fractions with target thickness we apply a very simple model. The measured charge state fractions are considered as a joint result of the following

few processes: multiple capture at the entrance surface, reionization and/or single capture inside the bulk, capture at the exit surface (see also Fig. 3), and possibly autoionizing decay of excited states between target and detector.

Radiative decay, which is dominant in the high-Z domain, does not change the final charge state. For instance, for a single K vacancy state in a neutral Pb atom the Auger rates are typically 10^{14}–10^{16} s^{-1}, whereas the radiative decay rates reach values even beyond 10^{17} s^{-1} [22]. As these rates depend on the number of available electrons (approximately quadratically and linearly for autoionizing and radiative decay, respectively), especially the Auger rates will be significantly reduced in our case of highly charged Pb ions. Hence we may assume for this case typical rates of 10^{14} s^{-1} and 10^{16} s^{-1} for Auger and radiative relaxation, respectively. A rate of 10^{16} s^{-1} will give a partial decay length of 10 nm which is comparable to the thickness of our thinnest target (about 45 nm). With our technique the influence of the autoionizing decay on the detected charge state distributions is not accessible directly but can be crudely estimated by extrapolating the measured data for single electron capture to zero target thickness. Indeed, from the possible offset a very small probability for autoionizing contributions may result: for instance, for the 46 $\mu g/cm^2$ targets this would mean a 4% contribution to single capture via initial multielectron capture. This small value together with the reasonable good agreement of the single capture cross sections with theory shows that our measured single capture probabilities are mainly due to single capture events in the bulk. We stress that single capture dominates all the possible multicapture processes that are all together always well below the few percent level (see Fig. 2).

As mentioned already, multiple capture events in single collisions along the complete depth of the target are expected to have a too small probability and should additionally result in a monotonous increase in yield with target thickness. The multiple electron capture probabilities can also not be explained by subsequent single capture collisions inside the bulk. If one takes into account reionization in the bulk (P_l, loss probability) a yield proportional to $\frac{P_c}{P_c+P_l}(1 - e^{-(P_c+P_l)x})$ is predicted. Thus the multicapture yield is expected to saturate but certainly not to decrease with increasing target thickness.

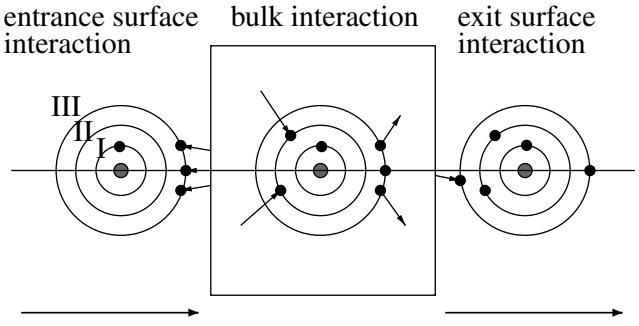

FIG. 3. Illustration of the three-state model (see text) for the key processes during the interaction of a highly charged ion with a solid target.

The observed target thickness dependences can be explained, however, if more electrons are captured when the ion enters the target, i.e., at the entrance surface. The multiple capture yield is then independent of the target thickness and the sharp decrease in probability from 11 to 19 $\mu g/cm^2$ target thickness is due to a very high cross section, in the Mb region, for reionization inside the bulk. This also means that at the surface electrons have to be captured into higher states with respect to single capture in the bulk, thus forming with a certain small probability a hollow ion. Hollow atom formation has been widely observed for light ions in the keV energy regime. For Pb^{81+} ions in the keV energy regime one would find with the overbarrier model [23] a critical distance of about 70 a.u. for the onset of electron capture with the electrons being captured in the $n = 65$ shell. However, for a 46 MeV/u Pb ion having a velocity of 41.5 a.u. this model is not applicable and an enhanced probability for many electron capture at the entrance surface has been unexpected so far.

Because all targets used are self-supporting, no difference between capture at the entrance surface and the exit surface should be expected. Electrons captured at the exit surface when the projectile has just left the target are not subject to collision induced reionization and thus add a constant probability to the measured data. Ignoring for the moment capture in the bulk one would thus expect the total capture probability to drop from a certain value for target thickness zero (corresponding to the sum of the probabilities at the two surfaces) to exactly half this value for infinitely thick targets (corresponding to the capture probability from the exit surface alone). As above, Auger decay behind the target may then slightly reduce the detected number of captured electrons independent of the target thickness. Taking into account capture in the bulk will result in an increasing multicapture detection probability with target thickness as successive single capture becomes important. Thus for equal capture probabilities at the entrance and exit surfaces the measured yield should never drop by more than a factor of 2 from zero target thickness to the minimum value.

As can be clearly seen in Fig. 2 the measured multiple electron capture probabilities drop significantly by more than a factor of 2. This indicates that the probabilities to capture electrons at the exit surface are appreciably smaller than the probabilities to capture at the entrance surface. One possible explanation to account for this strong difference might be a time dependent polarization of the target electron gas by the ion: Already at large distances the strong field of the approaching ion can pull on the target electrons towards the entrance surface leading there to a possible enhancement of the electron density. However, at the exit surface—considering the typical plasmon frequency of 10^{14} s^{-1}—the ion will have left the target

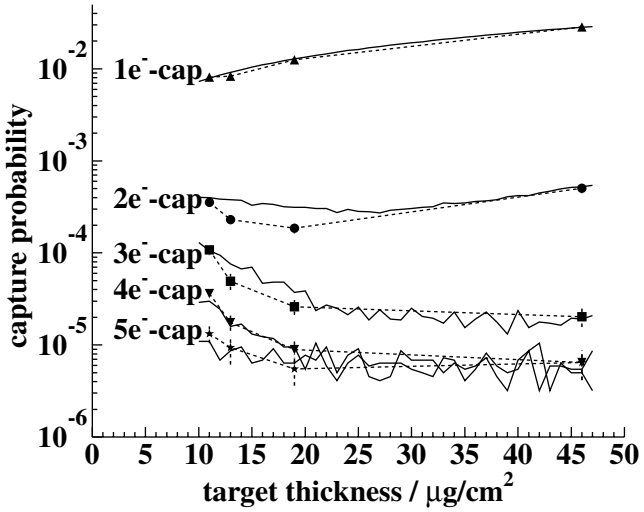

FIG. 4. Measured data (closed symbols with dashed lines) in comparison with a Monte Carlo simulation (solid line). The fluctuations in the Monte Carlo calculation indicate the statistical error of the simulation.

long before the electrons can react there and no further capture is possible.

A simple three-state model (see Fig. 3) can be applied with the initial electron being in the ground state I, electron capture in the bulk populating less tightly bound states II, and capture at the surface going into loosely bound states III. Within this crude model a Monte Carlo simulation was performed which reproduces our data quite well, as is shown in Fig. 4. Details will be published elsewhere [15]. In this simple simulation autoionizing decay is neglected for the reasons given above and the capture probabilities from the surfaces are taken to be the effective probabilities to detect the respective charge states. Capture in the bulk is treated as pure single capture with the measured cross section of 13.3 kb and a reionization cross section of 10 kb as discussed. Up to seven-electron capture at the entrance surface has been taken into account with a total (sum) probability of 1.5×10^{-3} for multiple capture, where the single electron capture probability is of the same order of 10^{-3}. This number and the probability distribution are adjusted to fit the data best with decreasing probabilities for the increasing number of electrons captured. The reionization cross section for states III is 1.8 Mb per electron, which has been extracted by an exponential fit to the measured data at thin targets. The probability of capture at the exit surface is taken as the difference between the measured probability and the bulk contribution ($P_1^n/n!$) at 46 μg/cm^2 target thickness; there, electrons captured at the entrance surface will have been reionized already. This gives multiple (and single) capture probabilities roughly an order of magnitude smaller than those at the entrance surface. This may point to a transient polarization of the target traversed by the heavy ion.

To conclude, we have for the first time studied the charge state distribution of highly charged, heavy ions at a moderate velocity traversing thin target foils. With hydrogenlike Pb ions at 46 MeV/u energy traversing thin carbon foils capture of up to five electrons could be observed, in particular, for the thinnest targets. The found target thickness dependence is in strong contrast to any expectation: The multielectron capture fraction increases drastically for thin targets with decreasing target thickness. We interpret this as evidence for a strong contribution of multiple electron capture at the entrance surface. In contrast, the corresponding capture probabilities at the exit surface seem to be 1 order of magnitude smaller. The simple three-state model developed describes our unexpected findings surprisingly well.

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