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Upper limit on the cross section for nuclear charge pickup by relativistic uranium ions

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We have searched for examples of nuclear charge pickup by relativistic uranium ions in targets of both uranium and phosphate glass. We find none, which allows us to set an upper limit of 7.7 mb per target atom at the 90% confidence level on the cross section for this process. An extrapolation of the approximately quadratic dependence on projectile charge of the cross section for charge pickup predicts a cross section which would be ~10 times larger. This breakdown in the scaling can be understood by the propensity of the actinides to fission upon the deposition of sufficient excitation energy.

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We have recently discovered that BP-1, a track-etch detector of remarkably high sensitivity which we developed several years ago [1], exhibits extremely good charge resolution, about $0.16e$, in measurements of uranium ions down to energies as low as 500 MeV u^{-1} [2]. Such resolution enables us to make direct measurements of the mean free paths for electron capture and loss in the glass detector [3], to measure cross sections for charge-changing fragmentation, and to search for nuclear charge pickup.

Our detectors consist of two stacks of BP-1 glass plates. The first stack was composed of two sheets of glass, followed by a uranium target, followed by several more sheets of glass. The second stack was composed of glass only. We exposed the two stacks to relativistic ^{238}U ions at the Bevalac at the Lawrence Berkeley Laboratory, the first to ions at 960 MeV u^{-1} and the second to ions at 920 MeV u^{-1}. The total fluence was ~15 000 and the incidence angle was 20° in each case. We etched the glass in 6.25 N NaOH at 40 °C for 20.3 h in the case of the first stack, and 19.4 h in the case of the second, for a total etched distance of 30.3 and 28.5 μm, respectively. We used our automated scanning system to measure the etch pit sizes by digitally fitting ellipses to the images of the mouths of the etch pits. The scanning system consists of a CCD camera attached to a microscope, an image processing system, and a computer. The system automatically locates, measures, and records the ellipse dimensions and orientations of ~10 000 etch pits per hour on a densely populated piece of glass.

The principle of operation of the track-etch detectors is described elsewhere [4]. Etching of the glass produces conical etch pits coincident with the point of penetration of the original ionizing particle into the detector; the size of the mouth of the etch pit is a sensitive function of the ionization rate of the original particle, which is to first order a function of $Z^\star/\beta$, where $Z^\star$ is the ionic charge of the projectile and β is its velocity. For a monoenergetic beam, the size of the mouth is related only to charge. Figure 1 is a histogram of the distribution of etch pit mouth sizes as measured at the top of the second stack. The charge resolution is about $0.16e$. The charge states of uranium in the fully stripped state and with one and two electrons attached are easily distinguished. The valleys between the peaks are partially filled in, due to charge-state changing within the sampling region of the glass, which is ~20 μm in depth per surface.

In the first stack, we measured the sizes of etch pits on the bottom of sheet 1 and the top of sheet 2, and on the bottom of sheet 4 and the top of sheet 5 which was the deepest pair of adjacent surfaces for which we had sufficiently good charge resolution. The sheets are labeled in the order in which they were placed *downstream* of the uranium target. We selected events which had the same ratio of major axis to minor axis as the main beam, in

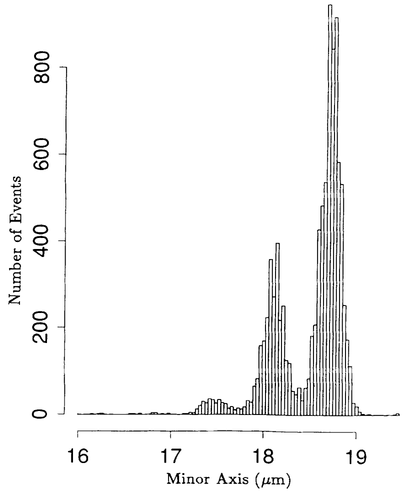

FIG. 1. Histogram of minor axes of etch pits as measured by the automated microscope system on the top of stack 2. The peaks correspond to ionic charge states of the uranium beam.

TABLE I. Charge pickup data.

Target	Thickness	Energy	$N(Z_{\rm app} = 92)$	$\sigma_{\Delta Z=+1}$	$\sigma_{\rm scaling}$
Air	139.7	951 − 960	3497	< 87.1	73.5
BP-1	0.20	911 − 951	3497	< 59.3	78.4
U	0.30	650 − 911	3497	< 45.4	109.7
BP-1	0.13	620 − 650	3497	< 91.3	78.4
BP-1	0.26	529 − 620	1773	< 90.0	78.4
BP-1	0.39	805 − 905	6201	< 17.2	78.4
all atoms				< 7.66	83.9

order to remove from the data set events which were doubled or had dust on the edge of the mouth. This cut is discussed in more detail in [10]. Because the thickness of the uranium target was slightly nonuniform, there were small variations in the amount of slowing experienced by projectile ions in passing through the target, which in turn led to small variations in $Z^\star/\beta$ across the detector. We therefore applied a small position-dependent correction to the data. There are no candidates for charge pickup in either pair of surfaces. We made measurements at two locations in this stack in order to improve the statistics. The number of events in the peak which corresponds to fully stripped uranium determines the statistical significance of a null result. The number of events in this peak is considerably smaller (see Table I) in sheets 4 and 5 than in sheets 1 and 2. This is due to nuclear charge-changing interactions in the intervening sheets and to the fact that the fraction of uranium projectiles in the fully stripped state is smaller downstream because of the reduced energy as discussed below.

In the case of the second stack, which consisted of glass only, we made similar measurements of etch pit sizes on the bottom of sheet 4 and on the top of sheet 5. We applied cuts to the data similar to those applied to the data from the first stack; in addition to these we applied cuts based on a scan of the bottom of sheet 2 to remove events from the dataset which entered the stack slower than the rest of the beam due to "beampipe-scraping" upstream. Figure 2 shows the correlation of measurements between matched etch pits on the bottom of sheet 4 and the top of sheet 5. Six charge states are evident, with charge-state changing events lying in the off-diagonal "boxes." Again, there are no candidates for charge pickup.

Because of the large probability for electron attachment in this charge regime, some fraction of a population of charge-pickup events would lie in the $Z^\star = 92$ or $Z^\star = 91$ peaks and so would be missed. Since these fractions will differ from the corresponding fraction for uranium by no more than 3% [5], we use the size of the peak which corresponds to fully stripped uranium for normalization of calculations of cross section.

We have three targets: air, uranium, and BP-1 glass. The main constituents of BP-1 are oxygen (63.1 mol%), phosphorus (17.1 mol%), and barium (10.9 mol%), with smaller amounts of sodium and silicon. Since the dependence of cross section for charge pickup on *target* charge for projectiles of gold ($Z = 79$) and lighter is found to be weak [6, 10], we treat all target atoms as the same in our analysis, and calculate the cross section *per atom*.

Guoxiao *et al.* [6] have found that for projectiles with energy of ~1 GeV $\rm u^{-1}$ the cross section for charge pickup varies approximately as the ***square*** of the projectile charge. This scaling is expressed by

$$\sigma_{\Delta Z=+1} = 1.7\times10^{-4}(A_{\rm proj}^{1/3} + A_{\rm targ}^{1/3} - 1)A_{\rm proj}^2\ \rm mb.$$

We show our results in Table I, along with the corresponding predictions of the scaling law derived in [6]. Figure 3 shows the previous data along with the new upper limit.

The scaling in [6] was derived from measurements made at ~1 GeV $\rm u^{-1}$. Binns *et al.* [8] and Guiru *et al.* [9] have found that the cross section for charge pickup increases with decreasing energy down to ~500 MeV $\rm u^{-1}$. This behavior has not been quantified, but it implies that our upper limit is even further below an ***energy-dependent*** extrapolation from lighter projectiles.

Here we investigate the possibility that the reduction in cross section that we observe is due to fission of the

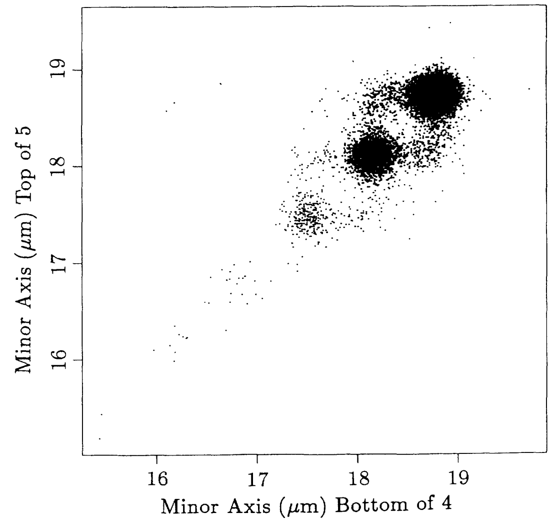

FIG. 2. Scatter plot of measurements of minor axes of etch pits on the bottom of sheet 4 and the top of sheet 5 of the second stack. These surfaces were adjacent during the exposure. Six charge states are evident: the three heavily populated regions are primarily ionic charge states of the uranium beam; the others are various ionic charge states of nuclear fragments. Off-diagonal events which are not found in the charge-switching "boxes" are probably due to mismeasurement caused by dust.

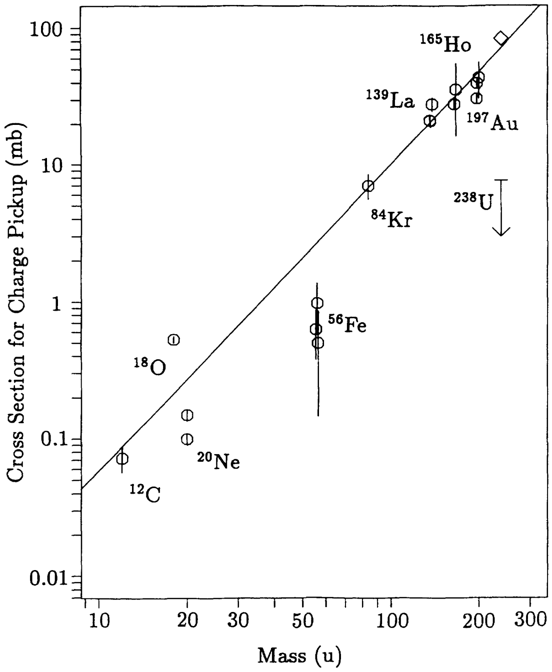

FIG. 3. Cross section for charge pickup as a function of projectile mass. Some points for ^{165}Ho projectiles are taken from [10], the upper limit for ^{238}U projectiles from the present paper, and the remaining points measured or referenced in [6]. The line is the best-fit power-law $\sigma_{\Delta Z=+1} = 1.7\times10^{-4}(A_{\text{proj}}^{1/3}+A_{\text{targ}}^{1/3}-1)A_{\text{proj}}^{2}$ mb from [6]; ◇ is the prediction for the current measurement based on this formula.

excited neptunium projectile after the charge-pickup interaction. If we take the actual cross section for charge pickup σ_{scaling} to be that predicted by the scaling law in [6], then the probability for fission p is

$$p = 1 - \frac{\sigma_{\text{obs}}}{\sigma_{\text{scaling}}},$$

where σ_{obs} is the observed cross section for pickup. From Table I we have $\sigma_{\text{obs}} < 7.66$ mb and $\sigma_{\text{scaling}} = 83.9$ mb, giving $p > 0.908$.

An excited actinide nucleus loses energy by neutron evaporation, each neutron carrying away 5–7 MeV, but the nucleus has a competing channel to fission. The competition between these channels is usually expressed by Γ_n/Γ_f, the ratio of the width for neutron emission to that for fission. Γ_n/Γ_f varies with isotope, excitation energy, and angular momentum, and can span many orders of magnitude [7]. However, the Γ_n/Γ_f varies only slowly with excitation energy for easily fissionable nuclei near the line of β stability. In fact, good fits have been obtained for proton-nucleus spallation reactions of up to 300 MeV assuming that Γ_n/Γ_f is constant ([7], p. 239). After each neutron emission, therefore, the hot nucleus has a roughly constant probability for fissioning instead of emitting another neutron. Thus, the probability for the hot nucleus to cool to the ground state without fissioning has a stair-step dependence on excitation energy.

Gavron *et al.* [11] have studied fission probabilities of $^{234-239}$Np in the reaction U(^{3}He,*df*) and U(^{3}He,*tf*). They found that the fission probability increases from ~0 at zero excitation energy to reach a maximum at about the neutron separation energy B_n, reaching almost 1 for ^{234}Np and ^{235}Np; from B_n up to ~12 MeV in excitation energy the probability for fission is roughly constant and lies between ~0.4 (for ^{239}Np) and ~0.8 (for ^{234}Np).

If we take the probability for fission to be constant at each stage of cooling, which is equivalent to the assumption that Γ_n/Γ_f is constant with energy and isotope, then

$$1 - p = \left(\frac{\Gamma_f}{\Gamma_n + \Gamma_f}\right)^{E_{\text{exc}}/E_{\text{evap}}}$$

or

$$E_{\text{exc}} = -E_{\text{evap}}\frac{\log(1-p)}{\log(1+\Gamma_f/\Gamma_n)},$$

where E_{exc} is the excitation energy and E_{evap} is the amount of excitation energy carried away by each evaporating neutron. We point out that the mass of the Np ion is unknown, since there may be prompt neutron emission during the charge-pickup process. For the projectile most likely to survive cooling, ^{239}Np, $\Gamma_n/\Gamma_f \sim 1.5$, so that $E_{\text{min}}/E_{\text{evap}} = 4.6$. Thus, it is clear from these data that an excitation energy no more than a factor of five higher than the neutron evaporation energy $E_{\text{evap}} \simeq 8$ MeV would be sufficient to ensure that >90.8% of the hot nuclei fission before they reach their ground states. That the projectile nucleus should have an excitation energy greater than 40 MeV after the charge-pickup process is likely. Thus the observed scaling violation can be understood in terms of the propensity of the actinides (in this case, neptunium) to fission upon the deposition of even a small amount of excitation energy.

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