Measurements of high energy loss rates of fast highly charged U ions channeled in thin silicon crystals

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The results of two channeling experiments show that highly charged heavy ions at moderate velocities ($v \ll Zv_0$) may lose more energy in the traversal of a thin crystal when they are injected along a major crystallographic direction than when they traverse the crystal in random conditions. This is due to the fact that the large reduction of electron capture probabilities allows them to keep their high electronic charge throughout the crystal, which is not the case for projectiles traveling in random conditions. Although channeled projectiles experience reduced electron densities, their energy loss rate, that is, at first order, proportional to the square of the ions charge, is then strongly enhanced. This feature could be used as a step for decelerating highly charged ions from the high energies that are needed to produce them, and also to improve our understanding of the slowing down of very highly charged projectiles at low velocities, for which the current perturbative models are not well suited.

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The interaction of fast positive projectiles with aligned crystals has been studied for more than 40 years.[1–3] In the particular case of positive heavy ions transmitted through a thin crystal in channeling conditions, this interaction may be characterized by the well-known following properties of the channeled projectiles: being prevented from approaching target atomic planes or strings, they experience lower target electron densities than projectiles traveling in random conditions. As a consequence, they usually suffer a reduced energy loss during the crystal traversal.[4–7] In the case of fast bare projectiles such as MeV protons or α particles, the reduction factor ε is close to 0.4 for the best channeled ions. However, the ability of fast heavy ions to lose electrons attached to them or to capture target electrons is also reduced: the channeled ions keep the memory of their charge state at incidence for longer times,[8] and then the charge distribution of the projectiles transmitted in channeling conditions is usually very different from the Gaussian-like distribution observed in random conditions (that most often reflects charge state equilibrium). In particular, it may happen that the best channeled projectiles stay frozen in their initial charge state throughout the crystal. If this charge state $Q_{\rm fr}$ is very different from the mean charge state at equilibrium Q_R of the ions of the same velocity range traveling in random conditions, the change in energy loss in channeling conditions is determined not only by the reduction of the mean encountered electron density, but also by the dependence of the mean energy loss ΔE upon the charge Q (ΔE is proportional to Q^2, at first order).

If $Q_{\rm fr} < Q_R$, the two effects add up, which results in a strong reduction of energy loss. For instance, in an experiment performed by our collaboration[9] with 27 MeV/u Xe^{35+} in a thin silicon crystal from which the random projectiles were emerging with a mean charge 50+, the energy loss of the small transmitted frozen 35+ fraction was measured to be 0.20 times the random energy loss, i.e. close to $\varepsilon(Q_{\rm fr}/Q_R)^2$.

If $Q_{\rm fr} > Q_R$, the two effects oppose each other, which can result in the paradoxical situation where channeled projectiles lose more energy than projectiles traversing the crystal in random conditions if $(Q_{\rm fr}/Q_R)^2$ is larger than $1/\varepsilon$.

This last case is the subject of the present paper, which was initially motivated by the idea that slowing down highly charged heavy ions without letting them capture electrons, i.e., by channeling them in a crystal, could be a useful stage in the process of stopping and trapping highly charged ions produced at a high velocity. Moreover, because the very strong perturbation induced on the target electron gas by the passage of slow, highly charged heavy ions cannot be described properly by the existing energy loss theories, the experimental determination of the stopping power for such ions is obviously interesting. Of course, the yield of frozen transmitted projectiles is expected to decrease rapidly when the crystal thickness increases since this decrease is favored by the transverse energy heating (dechanneling by multiple electron scattering) and also by a fast increase of all the capture cross sections when the ion velocity decreases.

We have performed two experiments at GSI (Darmstadt) with H-like U^{91+} ions. Uranium ions are accelerated up to several hundreds of MeV/u by the SIS synchrotron, stripped by passage through a thin foil, and the bare ions are injected into the experimental storage ring ESR. After being cooled and decelerated down to a few MeV, H-like ions are extracted by radiative recombination[10] and sent into the channeling beam line.

The first experiment has been already described in detail in a recent paper.[11] Briefly, a beam of $E_{\rm inc} = 20$ MeV/u U^{91+} ions (mass M, velocity v, charge Q) was sent onto a silicon single crystal in which the path length was 11.7 μm, and the transmitted projectiles were charge- and energy-analyzed by means of a magnetic spectrometer and collected into a two-dimensional (2D) position-sensitive detector located at the focal plane of the magnet. In channeling geometry, the contribution of each outcoming charge state could be separated, which was not the case for a random orientation. However, for the latter, the position of the centroid of the transmitted ion distribution nearly coincided with that of the outcoming charge state 74+, which was part of the unchanneled fraction of the beam observed in channeling geometry. This charge state is very close to the mean charge state at equilibrium Q_{eq} obtained with the semi-empirical formula proposed by Leon *et al*. (74.2+).[12] It is also possible to extract the value of Q_{eq} by assuming that the random energy loss corresponds to the value given by the SRIM code[13] and then to deduce the charge state associated with the centroid of the transmitted ion distribution. With this procedure, we find Q_{eq}=74.37+ for an outcoming energy of 19 MeV/u (SRIM predicts an overall loss of 1 MeV/u, i.e., 5% of the incident energy). We are thus led to conclude that, at this energy, Q_{eq} lies between 74+ and 74.5+. The fact that the individual contribution of the outcoming charge states could not be isolated in random geometry rules out the possibility to check in a reliable way the accuracy of the SRIM prediction for the stopping power of these highly charged heavy ions in an intermediate velocity domain. Finally it may be interesting to compare our result on Q_{eq} to what is predicted by other authors. For instance, the CASP code[14,15] gives a value of 75.6+ at 19 MeV/u, not too far from what we find. However, when using this value, the energy loss deduced from the position of the centroid of the outcoming ion distribution is found to be 62% lower than predicted by SRIM, which appears questionable. An even much higher value, $Q_{eq} = 79.76$ is proposed by Nicolaev and Dimitriev,[16] which, comparing to our results, seems unrealistic.

For incidence along the [110] axis direction, the channeled ions were able to keep very high charge states throughout the crystal, which could be identified individually. In particular, ∼25% of the incident beam was observed to be frozen in the incident 91+ charge state. A detailed analysis of the x-rays detected with a Ge detector, in coincidence with each charge-analyzed transmitted projectile, allowed us to evaluate the role of the various capture and deexcitation mechanisms taking place inside the crystal. In particular, radiative electron capture (REC) was observed to be the only capture process possible at distances above 0.6 Å from atomic strings.

The mean energy loss of the 91+ component observed for incidence along the [110] axis direction was 4.5% of E_{inc}, i.e., 90% of the corresponding random value. However, this large (25%) component of channeled ions corresponded to a rather wide distribution of transverse energies and then of energy losses for these ions, which experienced different electronic densities. Then we could determine that the best channeled ions had an energy loss of $0.038E_{inc}$, whereas the least well channeled 91+ ions had an energy loss of $0.053E_{inc}$, very close to the 1.0 MeV/u = $0.050E_{inc}$ value given by SRIM and corresponding to random conditions. Thus, for these ions, the ratio $(Q_{fr}/Q_R)^2$ was nearly equal to $1/\varepsilon$.

In the second experiment, for which we used essentially the same experimental setup, we sent 12 MeV/u U^{91+} ions onto a Si single crystal in which the path length was 18.3 μm. As the incident energy is lower than in our preceding experiment, the stopping cross sections and, mainly, the electron capture cross sections are higher. Moreover, because the crystal

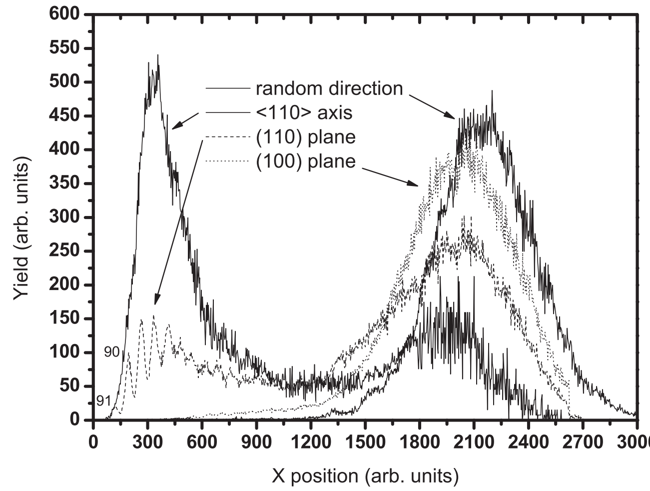

FIG. 1. X position distributions of transmitted U ions in the focal plane of the spectrometer, for four incidence conditions: random incidence and alignment along [110], (110), (100), directions, respectively. Incident ions: 12 MeV/u U^{91+}. Ion path length in the Si single crystal: 18.3 μm.

thickness has also been increased, the overall energy loss of the transmitted ions will obviously be much higher, and the probability for a channeled ion to be transmitted having kept its initial charge state will be very small. In fact, only some initially hyperchanneled ions are expected to be transmitted without having captured any electron. In order to minimize capture events in the disordered regions at the crystal entrance and, mostly at the crystal exit, we have tried to improve the surface condition by chemical cleaning (with HF acid) and passivation (with a hydrogen gas jet) before inserting the crystal into the vacuum chamber (10^{-7} Torr).

In Fig. 1 we show distributions corresponding to the positions of the transmitted ions in the focal plane of the magnetic spectrometer for four directions of incidence: in random conditions and for alignment along the (110), (100) planar and [110] axial directions. The four distributions, normalized to the same number of transmitted particles, correspond to projectile magnetic rigidities (Mv/Q) that increase from the left to the right. For incidence along a random direction, a large, essentially Gaussian-like peak is observed, which results from the smearing of the equilibrated charge distribution by energy loss straggling (mainly due to frequent ion charge changing in the target). As the position of the peak centroid results both from the mean charge state and from the energy at emergence, we had to assume a value for one of these two quantities to deduce the value of the other. The SRIM code[13] predicts an overall loss of 1.97 MeV/u. For this value, which corresponds to an exit energy of 10 MeV/u, we deduce from the peak position, Q_{eq} = 65.5+, which is again rather close from the prediction of Leon *et al.*[12] at this energy, Q_{eq} = 65.2+. Here also our experiment does not allow a precise check of the stopping power calculated using SRIM, which may be even more questionable at 10 MeV/u than at 20 MeV/u. We reach, however, a rather accurate estimation of Q_{eq}. For instance, when using the value given by the CASP code,[14,15] Q_{eq} = 67.9, we deduce from the peak position a stopping power that is 38% lower than found by SRIM, which is not very likely. The value $Q_{\mathrm{eq}} = 73.35$ proposed by Nicolaev and Dimitriev[16] is inconsistent with our measurements.

For incidence along the axial [110] direction the distribution splits into two parts: a small peak sitting nearly at the same location as the peak observed in random conditions, and a large peak corresponding either to higher charge states or to higher energy losses, or to both of them together.

It is clear that the first peak is due to the unchanneled part of the incident beam and to poorly channeled projectiles that are rapidly dechanneled. These projectiles can be expected to emerge with a mean charge state of 65+, as in random conditions, and the small shift with respect to the "random" peak can be attributed to the fact that the poorly channeled ions may keep a high charge state longer than in random conditions and then lose slightly more energy. The second peak is due to the large component of channeled projectiles that keep high charge states. We will come back to this peak later on, after discussing the (110) case.

The (110) planar incidence yields the most interesting distribution, again with two components, a broad one corresponding to the unchanneled and dechanneled ions, and the other corresponding to channeled ions, which show up with their individual charge states at emergence. The extreme left peak represents 0.3% of the incident beam, which means that it is composed of the very best channeled projectiles. In order to determine the charge states associated with this peak and with the following ones, we have studied the distributions obtained in coincidence with the x-ray photons detected by the Ge detector viewing the crystal target. We do not show here the x-ray spectra that are similar to those obtained with 20 MeV/u U^{91+} ions and that were presented in Ref. 11. In random conditions the spectrum is composed of uranium L and K photons that are mainly due to decay cascades following mechanical electron capture (MEC) events into high-n shells. In channeling conditions REC is available to channeled projectiles, for which the MEC process is reduced, and then the L and K lines are reduced, and the L- and K-REC lines become dominant. It is important to note that whatever the capture process, at least one photon is emitted and can be detected for the first capture event.

In order to illustrate how we could proceed to the peak identification, we present in Fig. 2 enlargements of the distributions of the transmitted projectiles corresponding to the high charge region. The distributions obtained in coincidence with the detection of a photon emitted under impact and without coincidence are both represented. Figure 2(a) corresponds to the distributions obtained for the (110) planar incidence: in the distribution measured in coincidence the far left peak has disappeared, which proves that this peak corresponds to frozen 91+ ions that had suffered neither electron excitation nor electron capture. In the figure, the height of the distribution obtained in coincidence, which contains much less events than the other one, has been multiplied by a factor $\boldsymbol{k}$ ($\gg$1). This factor has been chosen such that the 90+ peaks of the two distributions have the same height. The $\boldsymbol{k}$ value reflects essentially the detection solid angle. The probability for a photon following an electron capture by a 91+ ion to be detected is then $1/\boldsymbol{k}$. Thus, by subtracting the normalized coincidence distribution from the other one, one isolates the 91+ peak. Identifying this peak allows us to determine the corresponding most probable energy loss that we find equal to 2.59 MeV/u, i.e., 1.32 times the tabulated energy loss of ions traveling in random conditions.

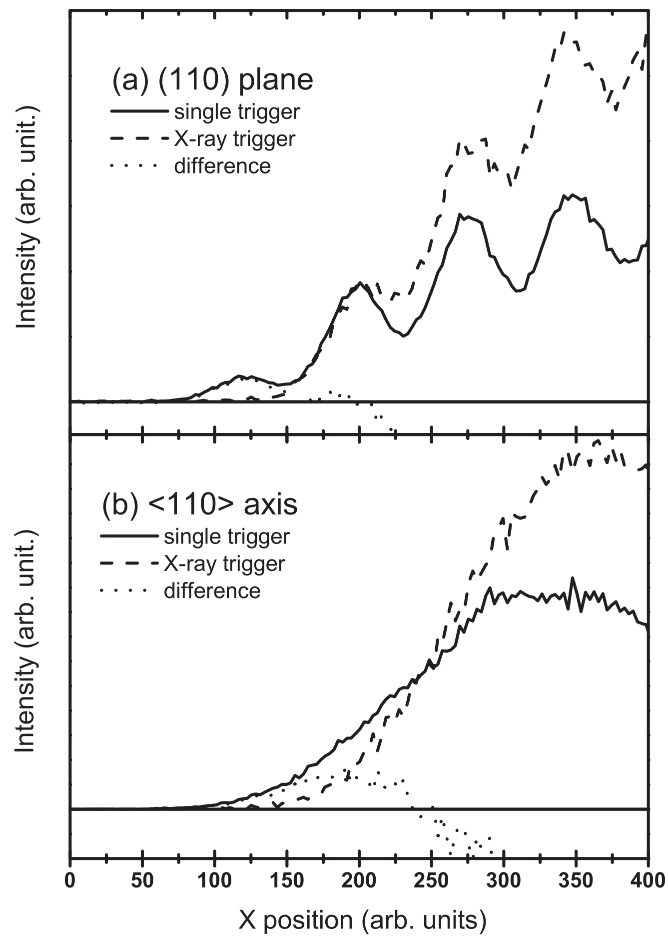

FIG. 2. Enlargements of the high-charge part of X position distributions for (a) (110) planar incidence and (b) [110] axial incidence: Distributions obtained without coincidence (single trigger), with coincidence (x-ray trigger), and their difference (see text).

Coming back to the distribution corresponding to the [110] axis, presented in Fig. 2(b), we apply again the procedure described above, subtracting the distribution obtained in coincidence with the detected x-rays from the other one, after multiplication by the same factor $\boldsymbol{k}$ as in the planar case. We felt allowed to keep $\boldsymbol{k}$ constant because we could show, in the experiment performed at 20 MeV/u, that the REC probabilities for emergent 90+ ions are equal for [110] and (110) incidences. Then, the subtraction should cancel the 90+ contribution allowing us to observe the 91+ peak isolated. This is indeed what happens: a wide peak remains, which is evaluated to represent ~1% of the total beam and corresponds to a mean energy loss of 2.11 MeV/u. This value is 1.07 times higher than what is predicted by SRIM in random conditions. The reason for the large peak width, as is probably the case for peaks corresponding to the lower charge states, thus preventing the contribution of individual charge states to show up in the distribution corresponding to the [110] direction, as they do in the (110) planar case, is not obvious. A possible explanation can be found if one observes Fig. 1, corresponding to the

(100) plane. The high charge state fraction transmitted along this plane is surprisingly small, reflecting a surprisingly small amount of well-channeled ions, even when considering that it is a less open planar channel than (110), and, as for the [110] axis, no peaks corresponding to individual charge states can be detected. In the geometry of our experiment, the (100) and (110) planes are, respectively, vertical and horizontal. We are then led to assume that the horizontal beam angular divergence of the incident beam is larger than the vertical one. Such differences have already been observed and measured during our first experiment with extracted beams from the storage ring.[3] Optimizing the focusing along the two directions would probably allow us to observe significantly higher fractions of transmitted frozen ions along the [110] and (100) directions, and probably also to improve the energy loss measurements.

Coming back to the slowing down of well channeled ions, we then observe that the energy loss of U^{91+} ions transmitted along the (110) plane, 2.34 MeV/u, is 1.1 times the value observed when these same ions are transmitted along the [110] axis, which is consistent with the higher mean electron density encountered by the best channeled projectiles along a planar direction. In the meantime, this higher electron density is responsible for more electron capture by REC, and the fraction of projectiles frozen in their initial charge state is lower. As the probability of remaining frozen decreases exponentially with the electron density encountered, while the stopping power varies much more slowly with the latter, it appears obvious that if one wants to keep some transmitted ions frozen after much higher losses, one should choose a major axial direction.

Our experimental results are summarized in Fig. 3, where our measured values of energy loss rates in channeling geometry are compared to the tabulated (SRIM) variation with the incident energy E of the energy loss rate of U ions traversing a Si target in a random condition. This latter curve presents a maximum at an energy of 6–7 MeV/u, which results from a combination of the energy dependence of the energy loss rate, when the projectiles are bare ions, and of the decrease at decreasing velocities of the ion charge, as they can keep increasingly more bound electrons inside matter. For the ion velocities throughout the maximum of dE/dx, the maximum energy transfer to inner-shell target electrons is well above their binding energies—there is no threshold effect. We also show in Fig. 3 what the variation of the energy loss rate would be if U ions would remain 91+ at any velocity in random conditions (dashed curve). As for our experimental results, the vertical bar at 20 MeV/u gives the precisely measured range of energy loss rates of the U^{91+} ions transmitted frozen in their initial charge state along the [110] direction. As already stated, in this experiment the fraction of frozen ions was large, ∼25%, and thus this energy loss range is associated to the broad range of electron densities experienced by different frozen channeled ions. Starting from this energy-loss rate interval, we have represented a band, extending toward lower energies, assuming a $(1/v^2)$ dependence for the energy loss rate of frozen ions. Of course, the $1/v^2$ law applies only in the high velocity regime where a perturbation approach can be used, and becomes questionable in the intermediate velocity regime, between 20 and 10 MeV/u corresponding to our experiments. It still allows us to estimate trends. The position of this band with respect to the dashed curve yields the reduction of the energy loss rates for frozen channeled projectiles experiencing reduced electron densities. The energy loss rates for frozen ions obtained at 12 MeV/u incident energy along the [110] and the (110) directions are also displayed in the same figure; the data point abscissae correspond to the mean ion energies in the target. For the (110) direction a vertical bar gives the extreme values of the energy loss rate associated to the 91+ peak [cf. Fig. 2(a)]. For the [110] direction, the vertical bar also corresponds to the range of energy loss rates suffered by frozen 91+ ions. Those are associated with the wide peak obtained by the subtraction procedure already detailed [cf. Fig. 2(b)]. As expected, these bars are located within the energy loss band interval defined from the 20 MeV/u results. However, as discussed above, the fraction of transmitted frozen ions is much smaller in this second experiment (∼1%), and thus this bar should in principle be much narrower than the band interval and should be located on the lower part of this band. The fact that this is not what we exactly observe is probably related to the rather large horizontal emittance of the incident beam.

Nevertheless, the extrapolation of the hachured band in Fig. 3 shows clearly that the slowing down rates of channeled U^{91+} ions reach high values at low energies, being, for example, more than twice the random slowing down rate for ∼5 MeV/u U ions. Of course, the probability for a U^{91+} ion to remain in this charge state is decreasing rapidly when it travels at low velocities: on the one hand, the REC cross sections increase for decreasing ion velocities, whereas the MEC capture becomes possible for larger impact parameters with respect to target atoms. Moreover, the transverse heating of channeled projectiles (i.e., the slow increase of their transverse energy as they proceed through the crystal) allows them to get closer to the crystal atomic strings, where electrons densities are higher, and then to increase their probability to suffer a MEC or REC event. This matter is partly illustrated

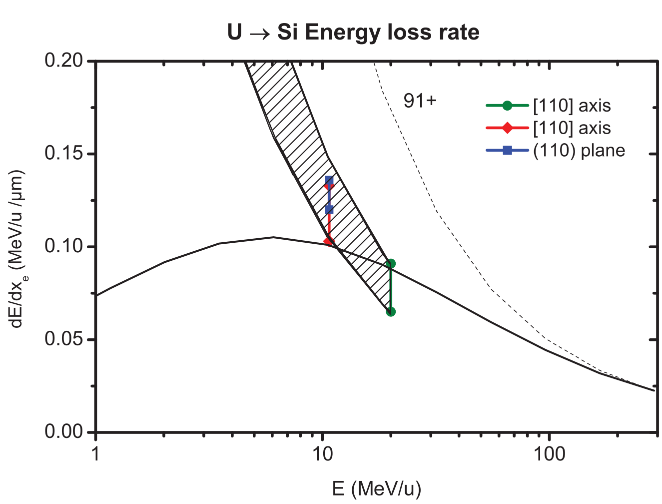

FIG. 3. (Color online) Energy loss rates of U ions in silicon. In random conditions: tabulate values (solid line) and values expected if the incident U ions could stay 91+ (dashed line). In channeling conditions: for 20 MeV/u incident ions, the vertical bar represents the range of energy-loss rates observed for the [110] incidence and yields the hachured band delimited by the two continuous lines (see text); for 12 MeV/u incident ions, the two bars represent the range of values measured for the [110] and the (110) incidences.

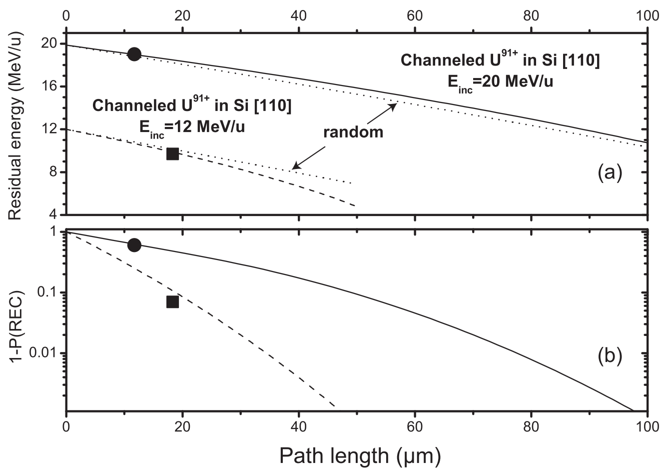

FIG. 4. (a) Calculated path length dependences of the residual energy for frozen U ions in the 91+ charge state when channeled along the [110] direction at 20 MeV/u (solid line) and 12 MeV/u (dashed line) and for ions transmitted in random conditions (dotted lines). (b) Calculated path length dependences of the surviving 91+ charge state fraction in channeling conditions for 20 MeV/u (solid line) and 12 MeV/u (dashed line).

in Fig. 4, where we show the role of the traversed crystal thickness on the 20 and 12 MeV/u U^{91+} ions incident along the [110] direction of a silicon crystal. In Fig. 4(a) we compare the residual energy of U ions after a given path length, in random and in channeling conditions; in the last case the path length is calculated assuming that the frozen U^{91+} channeled projectiles experience a mean electron density of 4 electrons/atom (2×10^{23} cm^{-3}), which is the density of valence electrons in silicon. From the map of mean electron densities along the [110] direction in silicon,[3] one can estimate that half of the channeled projectiles explore a region of the crystal where the local electron density is smaller than this value. However, 91+ ions at ∼10 MeV/u interact with valence, and even L-shell, Si electrons, at distances above 1 Å. Thus, we estimated that assuming a stopping power corresponding to a uniform electron gas with the density of the Si valence electrons provides a good order of magnitude. In Fig. 4(b) we show how the survival probability of U^{91+} ions decreases when increasing the path length. In this calculation we consider only REC electron capture, and disregard transverse heating effects and MEC capture, both which are expected to play a minor role with respect to REC. This hypothesis is supported by the x-ray spectra that were registered during the experiment performed with 12 MeV/u incident ions. These spectra, not shown here, demonstrate that REC is the only capture process leading to 90+ emerging ions. A comparison of Figs. 4(a) and 4(b) shows that frozen ions can survive when being slowed down considerably: For $E_{\mathrm{inc}} = 12$ MeV/u, one can expect, for example, a survival rate of $\sim10^{-2}$ for an energy loss corresponding to 30% of the initial ion energy.

To conclude, our experiments show that a significant fraction (of the order of 1%) of a U^{91+} beam extracted from a storage ring at energies of the order of 10 MeV/u can be transmitted frozen through a crystal in channeling geometry after losing ∼20% of its energy. The stopping power for these channeled frozen ions can be higher than that for random ions, and the lower the energy, the stronger the increase. Extrapolations of our results indicate that using thicker crystal targets should make it possible to transmit ∼0.01% of the incident beam frozen in its initial charge state at energies down to 5 MeV/u. This could be an interesting step toward trapping very slow highly charged heavy ions. It would also provide the opportunity to study in detail the interactions of such ions with solid matter, namely, the energy deposition and the induced material modifications, in a situation where the perturbation introduced by the ion is so strong that all existing models can be questioned.

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