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Stopping power measurements for 100-keV/u Cu ions in hydrogen and nitrogen

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Abstract

A new experimental area has been set up at the RFQ accelerator at ITEP (Moscow) to investigate the interaction of ions with gases and plasmas. The HIPr-ITEP accelerator provides ion beams at a specific energy of 100 keV/u. This energy is particularly suitable for studying the interaction of ions with cold and ionized matter below the maximum of the stopping power. As a first step, the new installation was used to measure the energy loss of copper ions in the nitrogen and hydrogen gases. Equilibrium stopping power values were determined and (as no previous experimental data for the Cu ions below the stopping power maximum existed) compared to the available semiempirical codes SRIM 2000, SRIM 2003, and with the new STIP code based on a clearly defined theoretical model which is now under development at ITEP.

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Keywords: Ion beam; Stopping power; Time-of-flight measurements; Beam transport model; Stopping force

1. Introduction

The stopping power of matter for fast charged particles, especially for heavy ions, has been the subject of theoretical and experimental studies for many years [1–3]. Investigations in this field continue to be actively pursued at present time as well [4,5]. Knowledge of the stopping powers and energy losses for different ion–target combinations is required for a wide variety of experiments in the nuclear and atomic physics, from the design of experimental apparatus to the data processing. Experimental information on deceleration of heavy ions in solids, gases, and in plasma targets has many applications in such areas as medicine, material science, accelerator and detector physics, and in target design for inertial confinement fusion.

On the other hand, investigation of the interaction of charged particles with matter is a fundamental physics problem in itself. At the design stage of our experiment for probing the stopping properties of the hydrogen plasma for heavy ions, we were immediately faced with a lack of experimental data for comparative analysis. A search through the literature [4,6] has revealed that the experimental data for low specific energies ($E < 500$ keV/u) are rather scarce for heavy ($Z > 6$) ions, and for many ion–target combinations there are no measurements at all. Hence, the calibration of the existing codes [7,8] against experimental data becomes problematic in this energy range.

An impressive work on systematization of the available stopping power data for gases has been reported in [9]. However, the authors of [9] performed their analysis only for specific energies $\gtrsim 0.5$ MeV/u. The main contribution to the experimental database in this energy range was made by the group that carried out its measurements on the accelerator facilities at IPN (Orsay) and GSI (Darmstadt)

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[10,11]. Recently, energy losses of the Ne, Mg and Na ions in the hydrogen gas in the specific energy range of 0.1–1.5 MeV/u were measured at the TRIUMF accelerator (Vancouver) [12]. These results partly filled the gap in the existing experimental database. A comparison of the measured values with the semi-empirical codes [7,8,13] has shown that in the vicinity of the Bragg peak the existing codes cannot predict the stopping power better than to an accuracy of ≃20%.

Having made the above mentioned survey, we decided to conduct the measurements of the stopping power of gases for 100 keV/u Cu ions at the existing ITEP RFQ accelerator HIPr [15]. Unfortunately, at present there is no possibility to do measurements for other values of the ion energy in this interesting region. According to our original idea, this present experiment was deemed as a first step in the envisaged program of comparative investigation of the Coulomb stopping of heavy ions in cold and ionized matter.

2. Experimental setup

The beam of 100 keV/u ^{63}Cu ions at charge states $q = +2, +3$ was delivered by the ITEP linac HIPr-1, which uses the MEVVA (MEtall Vapor Vacuum Arc) ion source [14] as the injector [15]. The linac can accelerate ions with the mass/charge ratios of up to $A/q = 60$ (in atomic units), providing a beam current of up to ≃10 mA. First of all, a new beam transport line for multicharged heavy ions from the 27 MHz ITEP RFQ accelerator to the investigated target has been designed and assembled. To do the measurements in gases, the same target configuration was used that had originally been designed for the energy loss measurements in a hydrogen plasma [16]. Two collinear quartz tubes, each of 79 mm in length and 6 mm in diameter, were filled with the studied gas. The 2-mm gap between the tubes contained the same gas. At both ends, the gas column was confined by tubular diaphragms of 10 mm in length and 1 mm in diameter, which allows a windowless penetration of the ion beam into a gas or plasma column. The overall view of the experimental setup is shown in Fig. 1.

The beam transport line was designed and assembled in accordance with the results of numerical simulations performed with the code TRACE-3D [17]. The vacuum pumping system for the beamline and for the diagnostic section behind the target was based on the TMN-500 and TMN-1500 vacuum pumps (pumping speeds 500 and 1500 l/s, respectively). To protect the high vacuum in the beamline from the target gas, differential pumping by a turbo-molecular pump with a pumping rate of 500 l/s was used. Beside protection from the target gas, this pumping system ensured a relatively fast changeover of gas in the target

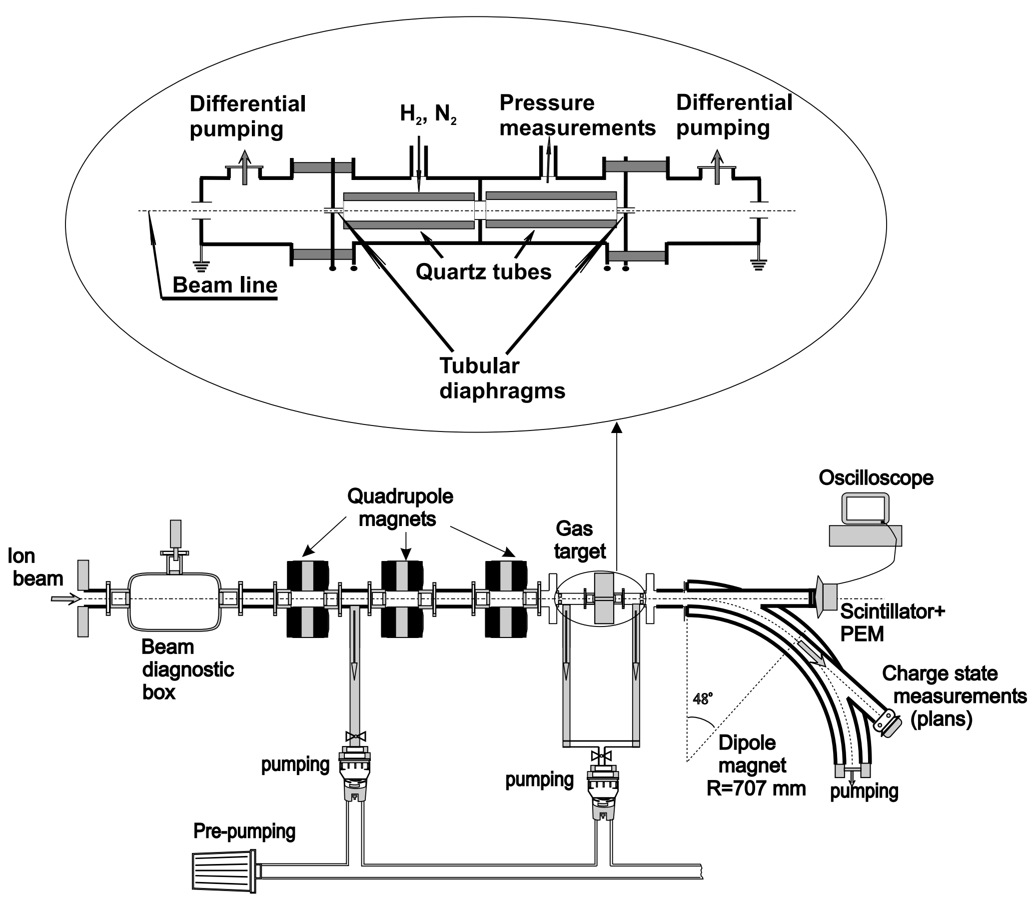

Fig. 1. Experimental setup for measurements of energy losses by low-energy heavy ions in gases and plasmas.

(according to our estimates, the full changeover of hydrogen occurred in less than 1 s). The test measurements have shown that a vacuum level of 1.5×10^{-6} mbar was secured in the beam transport channel when the gas (nitrogen) pressure in the target was 2 mbar. This allowed us to integrate the present target configuration into the acceleration tract.

The gas pressure inside the target was measured with a TPR-250 (Pfeiffer Vacuum) pressure gauge. The dynamic range of this gauge for the nitrogen gas is 10^{-3}–100 mbar, with an accuracy of $\sim$10% relative to the measured value. One of the shortcomings of this instrument is a weak and non-linear dependence of the output signal on the gas pressure when measurements are done in hydrogen. As a consequence, we were in fact unable to measure hydrogen pressures in excess of 1.6 mbar.

The target scheme did not allow us to control the gas pressure inside the tubular diaphragms. Hence, to evaluate the amount of gas along the tubular diaphragms at target ends, the gas density in this section was estimated on the basis of numerical simulations with the 1.5-dimensional hydrodynamics code [18], which had earlier proved to be quite efficient in simulations of expanding plasma targets. The gas flow was treated as one-dimensional along a tube with a variable cross-section, which decreased down the flow. At the exit end with a small cross-section, a boundary condition of exhaust into vacuum was applied. The opposite entrance end with a wide cross-section was treated in a simplified manner, as though the ideal gas there had a temperature T_0, density ρ_0, and only an axial velocity component. Such approximations result in a well-known steady-state flow for the efflux of a gas trough a converging tube: the gas density becomes constant along the narrow section of the tube with a fixed cross-section, where it has a value of

$$\rho = \rho_0 \left(\frac{2}{\gamma + 1}\right)^{1/(\gamma-1)} = 0.63\rho_0, \tag{1}$$

here $\gamma = 1.4$ is the adiabatic index of a two-atomic molecular gas. With the above estimate taken into account, we obtain the following value for the full effective length of the gas column along the beam path

$$L = 2 \times 79\ \text{mm} + 2\ \text{mm} + 2 \times 6.3\ \text{mm} = 172.6\ \text{mm}. \tag{2}$$

Time-of-flight measurements have been performed to determine the energy losses of ions. More specifically, we measured the phase shift in the periodic beam structure, i.e. the time delay of each ion bunch with respect to its initial (unperturbed) position in the temporal sequence. The measured time delay yields directly the change of the ion velocity with respect to its initial value. As a fiducial, the high-frequency signal from the master generator of the HIPr accelerator was used. To determine the energy losses in gas, two series of measurements were needed. In the first series, ions were transported through the empty (without gas) target under full vacuum; for the second one, the target was filled with gas. By measuring the time delays between the detected and the fiducial signals in the two series, the energy loss in the gas was obtained.

For practical realization of the above described method a registration scheme was chosen that combined a fast plastic scintillator NE102 with a photomultiplier FEU-87 (manufactured by Moscow electric-bulb factory). After traversing the target, the arrival time of the ion bunches was registered by the scintillator-based stop-detector; the obtained signal was digitized and recorded with a 1 GHz bandwidth oscilloscope at a sampling rate of 2.5 GHz, each record lasting 4 μs. Despite various measures taken to reduce noise pickup, no better than a 3:1 signal-to-noise ratio could be achieved. The noise induced by the master high-frequency generator had the frequency that was a multiple of the ion signal frequency. The width of the micropulses at half-maximum was $\approx$15 ns, whereas the shape of the individual bunches exhibited significant variations even within a single macropulse due to peculiarities of the generation and acceleration of the ion beams at the HIPr installation.

With the above mentioned facts in mind, Fourier analysis was performed at the first stage of data processing, and a spectral interval was isolated in the vicinity of the accelerator driving frequency ($\simeq$27 MHz). As a result, for each record (4 μs, $\approx$100 bunches) the mean shift (time delay) of the measured signal peak was determined with respect to its "prototype" in the fiducial signal from the high-frequency master generator. In this case, the principal error in the inferred value of the energy loss was due to the scatter in the phase shifts of the ion bunches with respect to the peaks of the fiducial signal. The above mentioned shape instability of the ion pulses produces an error of 0.7–1.2 ns in the position of the measured signal peaks, depending on the specific experimental run conditions (temperature, humidity, etc.).

3. Results of measurements

In the first place, the energy losses of 100-keV/u copper ions were measured in cold nitrogen gas. Generally nitrogen causes less load on the pumping system for the beam transport line than hydrogen, which makes it a natural choice for testing the overall installation performance. However, because of scarcity of experimental information in this energy range, the data obtained are also of their own interest. The energy losses in nitrogen were measured at four different pressure values ranging from 1 to 2.5 mbar. The results obtained are shown in Fig. 2. Although several series of measurements have been carried out, the errors in the energy loss values could not be noticeably reduced because of the scatter in the experimental data.

As the next step, the energy losses in the hydrogen gas at pressures between 1 and 1.5 mbar were measured. The lower pressure limit was determined by the sensitivity threshold for detection of the ion energy loss. On the other hand, in this experiment we could not increase the hydrogen pressure above 1.5 mbar when the gas was admitted into the target at a constant rate because the ion pumps,

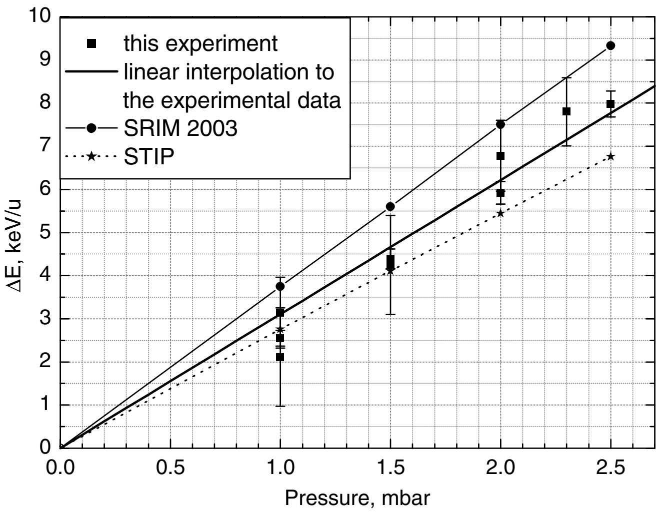

Fig. 2. Energy losses of 100.8-keV/u copper ions in cold nitrogen gas; the effective length of the gas cell was 17.26 cm.

used to maintain high vacuum in the acceleration tract, were not suited for pumping hydrogen and tended to break down after an extended period of hydrogen load. A possible solution to this problem in the future might be an introduction of fast shutters [19] that would open up only for the time of beam passage, which would reduce significantly the load on the pumping system of the HIPr accelerator.

The experimental results for the hydrogen target are given in Fig. 3. These data represent several series of measurements that have been carried out.

Because of relatively low values of the decrement ΔE of the ion specific energy as compared to the initial value of $E_0 = 100$ keV/u, the target may be considered as a thin one at all gas pressures. In this case the specific energy decrement is directly proportional to the gas pressure, and the slope of the corresponding straight line yields the stopping power of the corresponding gas at the initial ion energy. In this way, a linear interpolation to the measured dependence of ΔE versus gas pressure allowed us to obtain the following values of the stopping power of nitrogen and hydrogen for the 100 keV/u ions of copper

$$\begin{aligned} S_{\mathrm{H,exp}} &= 27.1 \pm 5.2 \text{ MeV cm}^2/\text{mg}, \\ S_{\mathrm{N,exp}} &= 9.9 \pm 0.6 \text{ MeV cm}^2/\text{mg}. \end{aligned} \tag{3}$$

The error bars for each individual measurement are rather large. However, by combining several series of measurements and assuming a linear dependence on the gas pressure, we were able to determine the stopping power of nitrogen to an accuracy of 6%, and the stopping power of hydrogen – to an accuracy of 19%.

4. Comparison with theory: the STIP code

New experimental data on the stopping power for fast ions are traditionally compared with the values predicted by the publicly available SRIM code [7]. We also present such a comparison in Figs. 2–4. Fig. 4 shows that the ion specific energy $E = 100$ keV/u is well below the Bragg peak on the $S(E)$ curve for Cu ions, i.e. in a region which is most difficult for theoretical modelling. A significant (by about 30%) discrepancy between the 2000 and 2003 SRIM versions indicates that the SRIM predictions may also be not very reliable in this region – which, in particular, keeps motivation high for conducting new stopping experiments with high-Z projectiles at low energies. As it is seen in Figs. 2 and 3, our experimental values of S for both the H_2 and N_2 targets are about 20% below the SRIM 2003 values (and about a factor 1.5 below the SRIM 2000 values).

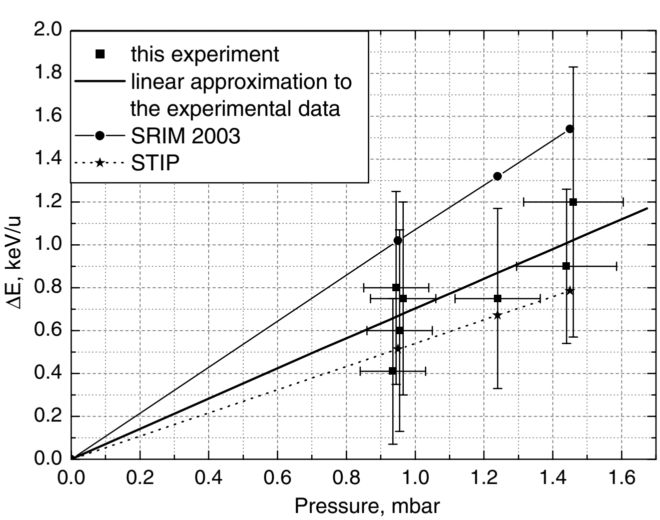

Fig. 3. Energy losses of 100.8-keV/u copper ions in cold hydrogen gas. The points at 0.95 mbar and 1.45 mbar are artificially slightly moved apart to ease the perception.

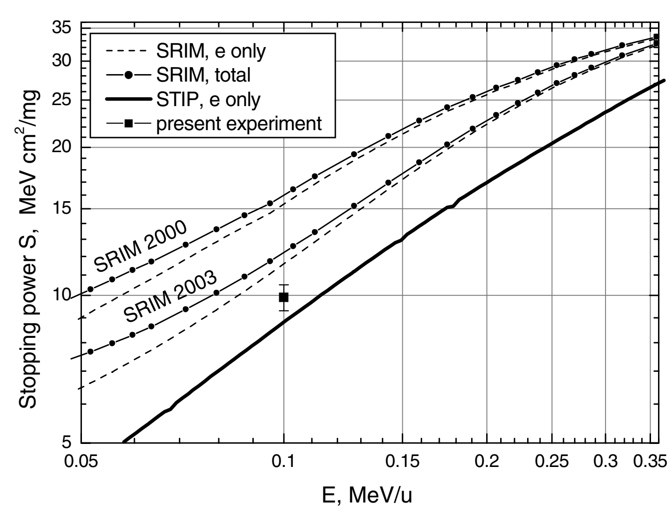

Fig. 4. Stopping power for Cu ions in nitrogen as a function of the projectile specific energy around 100 keV/u. Curves show the values calculated by the SRIM 2000, SRIM 2003, and the STIP codes for a Cu beam with equilibrium distribution over projectile charge states. For the SRIM code both the total (nuclear + electronic) and electronic (e-only) stopping powers are given; the STIP code does not take into account nuclear stopping.

The same tendency was noted in [12] for the Ne, Na, and Mg ions in a H_2 target.

Besides the semi-empirical SRIM model, based essentially on the concept of effective charge for heavy ions [7], a comparison with a clearly defined theoretical model would be of particular interest. As such, Figs. 2–4 show the values calculated with a new beam transport code STIP, which is currently under development at ITEP. The beam transport part of the STIP code is based on solving numerically the system of equations

$$\frac{\partial f_q}{\partial x} - \frac{\partial}{\partial E}(F_q f_q) - \frac{\partial^2}{\partial E^2}(D_q f_q) \\ = -(\kappa_{q,q+1} + \kappa_{q,q-1}) f_q + \kappa_{q-1,q} f_{q-1} + \kappa_{q+1,q} f_{q+1}, \quad (4)$$

for the distribution function $f_q(x,E)$ of the beam ions over the ion energy E and charge states $q = 0,1,2,\ldots,Z_p$ along the beam trajectory x; here $F_q = F_q(E) > 0$ is the stopping force acting on a projectile ion with a charge $+q$ and an energy E, $D_q = D_q(E)$ is the diffusion coefficient along the energy axis, $\kappa_{q,q\pm1} = n_e \sigma_{q,q\pm1}$ [cm^{-1}] are the rate coefficients, and $\sigma_{q,q\pm1}$ are the corresponding cross-sections for charge transfer events $q \to q \pm 1$. In this approach we abandon the notion of the effective charge and calculate explicitly the evolution of the projectile charge q due to single-electron events $q \to q \pm 1$. Energy loss associated with these charge transfer events is ignored. The diffusion term on the left-hand side of Eq. (4) accounts for the collisional energy straggling in the Fokker–Planck approximation; for the diffusion coefficients D_q the classical Bohr value [20] is used.

In addition to the beam transport model, the key ingredients in the STIP code are (i) a theoretical model for the stopping force $F_q(E)$ and (ii) appropriate formulae for the charge transfer cross-sections $\sigma_{q,q\pm1}(E)$ for which usually either a semi-empirical model or a fit to experimental data are used. The stopping force $F_q(E)$ is calculated by applying the Bloch [21] formula for a point-like charge $+q$ to each nl shell of the target atom, and then summing up the contributions from all shells. Where no atomic calculations are available, the mean excitation energies $\hbar\bar{\omega}_{nl}$ of individual shells, needed in the Bloch formula, are evaluated with an approximate semi-empirical procedure proposed earlier in [22]. Extension of the Bloch formula into the low-velocity region, where the stopping number (Coulomb logarithm) for a given atomic shell $L_{nl} \lesssim 1$, is done by using the formula (40) from [23], which represents the rigorous low-velocity solution for the classical Bohr model of harmonically bound target electrons. In this way we effectively account for the shell corrections – also in the region where they are large, i.e. where the Bloch formula formally leads to $L_{nl} < 0$. So far, contribution to the stopping power due to the collisions with target nuclei (nuclear stopping) has been ignored.

Under the conditions of the present experiment, charge changing events for the Cu^{q+} projectiles are dominated by (i) ionization in collisions with neutral atoms of the target gas and (ii) recombination via capture of bound electrons from the target atoms. For the ionization cross-section we used a generalization of the scaling law from [24], which takes into account a Thomas–Fermi-like screening of the nuclear potential by the bound electrons in target atoms. For the recombination cross-section, a semi-empirical formula from [25] was used. Simulations with the STIP code have shown that the equilibrium charge distribution of Cu projectiles, shown in Fig. 5, is established very rapidly, over a fraction of 1 cm of the target length even for the lowest pressure of 1 mbar in both the N_2 and H_2 targets. In particular, this implies that precise knowledge of the entrance charge (Cu^{2+} or Cu^{3+}) is actually irrelevant for our stopping power measurements.

In Figs. 2–4 one sees that the STIP values of the beam stopping power agree fairly well (within 15%) with the present experimental results but, in contrast to the SRIM values, lie systematically below the experimental points. The agreement becomes even better after one takes into account that the STIP results correspond to the electronic stopping only. As one can infer from the SRIM data in Figs. 2 and 3, nuclear stopping should add some 5–6% to the pure electronic stopping force from the STIP calculations.

It should be noted however that a good agreement between the STIP simulations and the experimental values may, to a certain extent, be spurious. The fact is that the present STIP model for $F_q(E)$ does not account for two important effects, namely, that (i) partially ionized projectile ions are not point-like charges, and (ii) the low-energy extension of the Bloch formula would be more appropriate to do on the basis of the quantum rather than classical oscillator model [23]. These two effects should, however, partially cancel one another [26], and their relative contribution remains to be investigated. It should be stressed also that an adequate description of the low-velocity regime of the Coulomb stopping is very important for the conditions of the present experiment: if we write the original Bloch

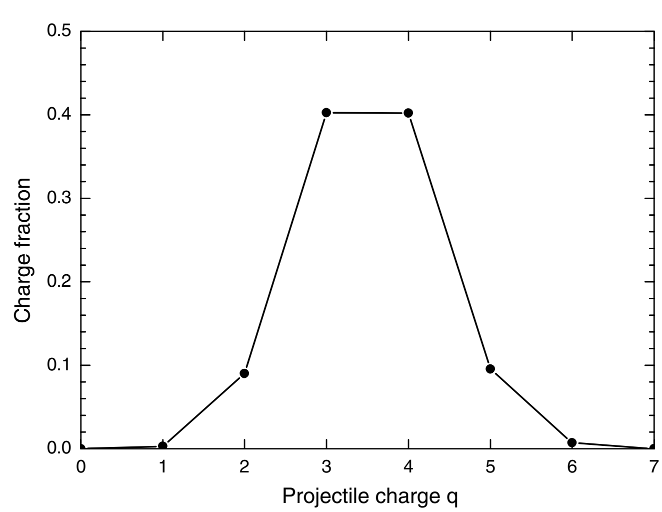

Fig. 5. Distribution of Cu ions over the charge states q, as calculated by the STIP code, after they traverse 1 cm of N_2 gas at a pressure of 1 mbar.

value of L_{nl} as $L_{nl} = \ln \Lambda_{nl}$, we find that $\Lambda_{nl} \approx 0.17/(\hbar\bar{\omega})_{\rm eV} \lesssim 0.01$ for 100 keV/u Cu^{3+} ions in our H_2 and N_2 targets.

5. Conclusion

New measurements of the stopping power of the nitrogen and hydrogen gases for 100 keV/u copper ions are reported. The main motivation for presenting these data stems, on the one hand, from general scarcity of experimental results near this low ion energy (which is already below the stopping power maximum for the copper ions) and, on the other hand, from the difficulties in theoretical modeling of the Coulomb stopping in this energy range. The latter, in particular, is reflected in a significant (by about 30%) discrepancy between the predictions of the 2000 and 2003 versions of the semi-empirical SRIM code. Our measurements clearly favour the SRIM-2003 values. A good (within 10–15%) agreement of the experimental results with the predictions of the newly developed STIP code indicates that a theoretical model, based on the charge-state approach combined with an adequate extension of the Bohr–Bethe–Bloch formula to low projectile velocities, may have good prospects for describing the Coulomb stopping of heavy ions near the stopping power maximum.

Acknowledgements

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