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Improved charge-state formulas

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Abstract

A large set of experimental charge-state distributions is analyzed in this work. Two fit formulas are presented for mean equilibrium charge states of projectiles ranging from protons to uranium. One formula is given for all ions in gas targets and another one for solid targets. The deviation from the experimental data is reduced by roughly a factor of two in comparison with the widely used formula by Nikolaev and Dmitriev as well as with the Bohr stripping criterion as revised by Northcliffe. Finally, the influence of the projectile charge state on the prediction of stopping powers for fast projectiles in carbon is shown and comparison is made with experimental energy-loss data. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

The prediction of ion-charge states in gases and solids is of importance for gas and foil strippers in accelerator systems as well as for the determination of projectile and target related quantities in ion–solid interactions. Especially the calculation of ion stopping powers, target excitation and subsequent materials modifications is largely influenced by the projectile charge state. For highly charged light ions most quantities, such as, e.g., energy loss and target-excitation probabilities, scale with the square of the projectile charge. Hence, uncertainties in the mean projectile charge are often magnified by a factor of two in the final results.

Well-known reviews on projectile charge states have been written by Allison [1] as well as by Betz [2]. Extensive tabulations of mean projectile charge states as well as shape parameters of the distributions are also available [1–5]. These tabulations and the corresponding original papers constitute the basis of the present work. The projectile charge state of fast moving ions in matter is mainly determined by the balance between electron capture and projectile–electron loss. Complications evolve from the influence of multi-electron loss processes, meta-stable excited states as well as Auger and radiative transitions of the highly excited projectile–electron system. The mean charge state in thin gas targets, however, may be derived with

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reasonable accuracy from (experimental) atomic physics data for the corresponding charge changing cross-sections [2,6].

In solids one observes an increased mean charge of fast ions. This is mainly due to the high collision frequency which exceeds the frequency of Auger and radiative decays. Thus, electrons in excited states are stripped off before they may decay to the ground state. Nikolaev and Dmitriev have derived semi-empirical charge-state formulas for fast heavy ions in carbon [7], which yields good results as long as the number of bound projectile electrons is not too low. A few single-electron models exist for the solution of the rate equations that govern the evolution of the projectile-state population. Only one code is known to us that allows to treat projectiles with up to about nine electrons [8,9].

In this work, we determine improved charge-state fits for projectiles ranging from protons to uranium. One formula is given for gas targets (fitted to experimental data mainly in the range of target nuclear charges from $Z_t = 1$ to 54) and another one for solid targets (fitted to experimental data for Be to Bi targets). Finally, the influence of the projectile charge state on the prediction of stopping powers for fast projectiles in carbon is shown and discussed.

2. Analysis of gas-target data

The set of experimental charge-state data for gas targets consists of 552 data points for the mean projectile charge-state q_{mean}. It stems mainly from [1,2] and the work cited therein. The measured mean equilibrium charge-state is subject to different sources of errors. These are the beam energy-calibration, statistical uncertainties (noise and background), slit scattering and the determination of an equilibrium thickness of the gas target. Comparison of measurements for similar projectile–target combinations in different laboratories indicates that the latter point limits the final accuracy to about ±0.4 charge states for heavy ions in gases.

A multi-parameter least-squares fit has been performed to find a reduced parameter x that minimizes the scatter of q_{mean}/Z_p (Z_p is the projectile nuclear charge) around a smooth curve which is part of the fit. The physical quantities that enter this fit are Z_p, the target nuclear charge Z_t, and the projectile velocity v_p divided by v_0 (v_0 is the Bohr velocity of 2.19×10^6 m/s). The result is

$$q_{\mathrm{mean}} = Z_p \frac{376x + x^6}{1428 - 1206x^{0.5} + 690x + x^6}, \tag{1}$$

with

$$x = \left(v_p/v_0 Z_p^{-0.52} Z_t^{0.03-0.017Z_p^{-0.52}v_p/v_0} \right)^{1+0.4/Z_p}. \tag{2}$$

The resulting fit is shown in Fig. 1 together with the experimental data for Z_p in the range 1–92 and Z_t in the range 1–54. Note that we have excluded ions below a certain threshold velocity. For these ions v_p as well as q_{mean} are low enough that the details of the target and projectile shell structure come into play. The corresponding resonant electron-capture processes depend significantly on the target–projectile combination, which prohibits any simple scaling behavior. These shell-structure effects appear to be extraordinary strong for the collision system He + He which has the highest ionization potential of all atoms. Therefore, only fast projectiles with $v > 2.8v_0$ have been included for this case.

The absolute uncertainty of the present charge-state fit is $\Delta q_{\mathrm{mean}} = 0.48$ and the relative uncertainty is $\Delta q_{\mathrm{mean}}/Z_p = 2.6\%$. This may be compared with $\Delta q_{\mathrm{mean}}/Z_p = 4.7\%$, the uncertainty of the Bohr stripping criterion [10,11] (revised by Northcliffe) when determined from the same experimental data set for $q_{\mathrm{mean}}^{\mathrm{experiment}}$. It is emphasized that we have found no remaining systematic single-parameter dependence when plotting $q_{\mathrm{mean}}^{\mathrm{experiment}} - q_{\mathrm{mean}}^{\mathrm{fit}}$ versus v_p, Z_p, Z_t, or versus the mean number of bound projectile electrons $Z_p - q_{\mathrm{mean}}$. A comparison of rare gases and diatomic molecules shows also no significant effect, although clear effects are visible in the charge-state *distribution* for specific projectile species and energies [2]. In fact, the uncertainty Δq_{mean} for the present fit appears to be consistent with the deviation of experimental data taken in different laboratories.

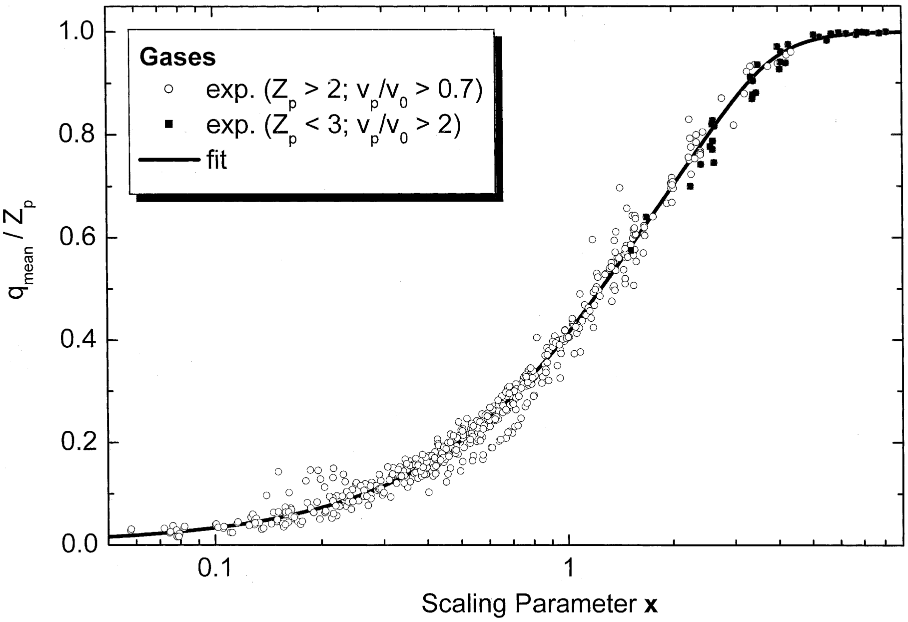

Fig. 1. Mean projectile charge-state as function of the reduced scaling variable x (defined by Eq. (2)) for gas targets.

3. Analysis of solid-state target data

A similar multi-parameter least-squares fit has also been applied to the solid-state data for q_{mean}. About 850 experimental data points have been accounted for and the estimated deviation between results of different laboratories amounts only to about ±0.2 charge states. The result of the fit is

$$q_{\mathrm{mean}} = Z_{\mathrm{p}} \frac{12x + x^4}{0.07/x + 6 + 0.3x^{0.5} + 10.37x + x^4}, \qquad (3)$$

with

$$x = \left(v_{\mathrm{p}}/v_0 Z_{\mathrm{p}}^{-0.52} Z_{\mathrm{t}}^{-0.019 Z_{\mathrm{p}}^{-0.52} v_{\mathrm{p}}/v_0} / 1.68 \right)^{1+1.8/Z_{\mathrm{p}}}. \qquad (4)$$

The corresponding fit curve is shown in Fig. 2 together with the experimental data for Z_{p} in the range 1–92 and Z_{t} in the range 4–83 (42% of the data correspond to carbon targets). Note that we have again included only ions above a certain threshold velocity. The absolute uncertainty of this charge-state fit for solids is $\Delta q_{\mathrm{mean}} = 0.54$ and the relative uncertainty is $\Delta q_{\mathrm{mean}}/Z_{\mathrm{p}} = 2.3\%$. This may be compared with the uncertainty of $\Delta q_{\mathrm{mean}}/Z_{\mathrm{p}} = 3.3\%$ corresponding to the well-known fit formula by Nikolaev and Dmitriev [7] when applied only to the subset of heavy ions with $Z_{\mathrm{p}} > 10$ in carbon. The uncertainty of that fit exceeds 10% for $\Delta q_{\mathrm{mean}}/Z_{\mathrm{p}}$, if light ions such as protons and helium are considered. Thus, the current fit is a significant improvement over the one by Nikolaev and Dmitriev.

Comparison of the uncertainties of Eqs. (3) and (4) with our estimate of the experimental uncertainties indicates that other “hidden” parameters influence $q_{\mathrm{mean}}^{\mathrm{experiment}}$. We have found no remaining systematic single-parameter dependence, except for $q_{\mathrm{mean}}^{\mathrm{experiment}} - q_{\mathrm{mean}}^{\mathrm{fit}}$ when plotted versus the mean number of bound projectile electrons $N_{\mathrm{b}} = Z_{\mathrm{p}} - q_{\mathrm{mean}}$. This dependence is shown in Fig. 3. Clear projectile-shell effects with pronounced maxima for 11 and 30 electrons are visible. These numbers are near to closed-shell configurations (10 for L shell and 28 for the M shell). Shell effects were found for the first time by Moak et al. [12,13] in the shape of charge distributions and later Shima et al. [4,5] have shown that the mean charge state in carbon targets is also affected. Fig. 3, however, allows for the first time to extract the absolute

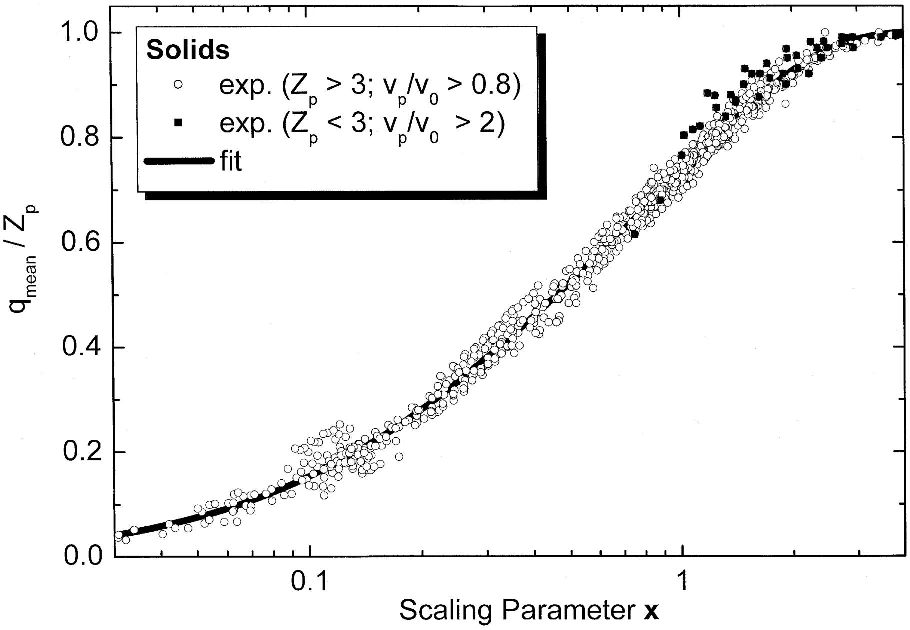

Fig. 2. Mean projectile charge-state as function of the reduced scaling variable x (defined by Eq. (4)) for solid-state targets.

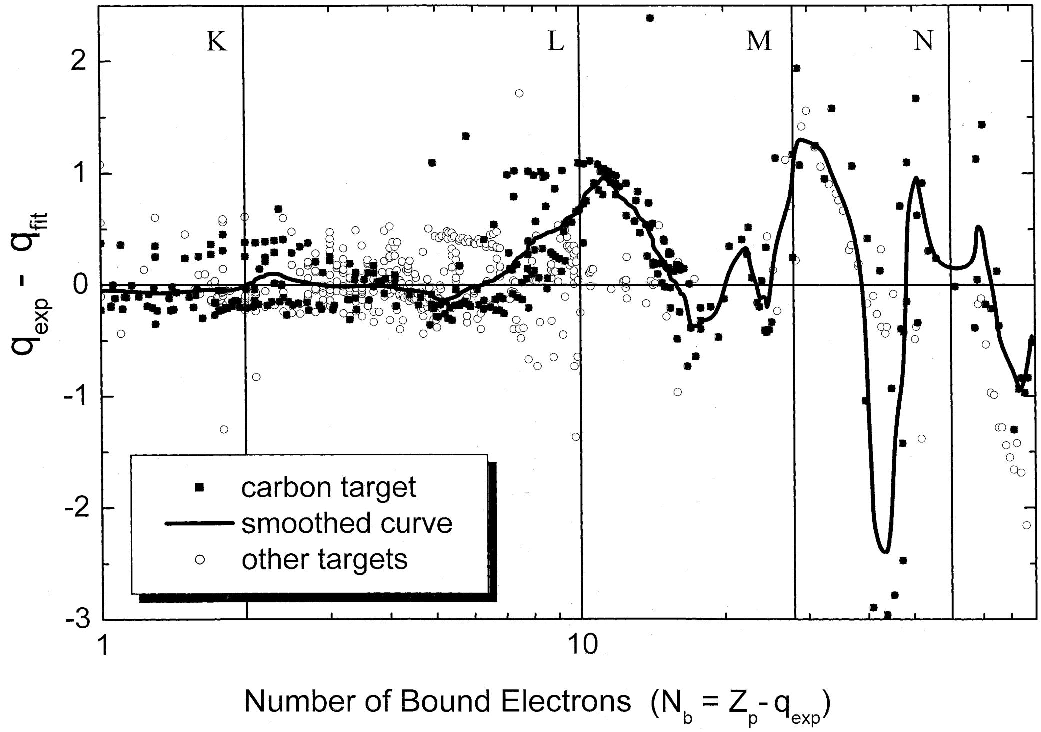

Fig. 3. Deviation of the mean projectile charge-state from the smooth fit curve (shown in Fig. 2) as function of the number of bound electrons for solid-state targets.

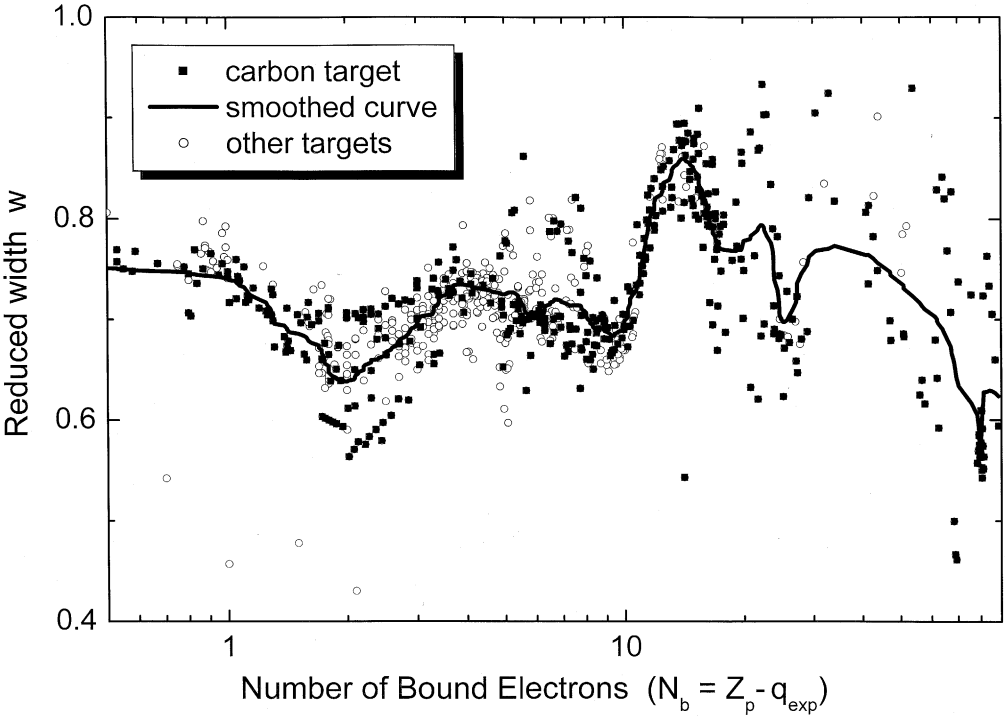

Fig. 4. Scaled width (see Eqs. (5) and (6)) of the projectile charge-state distribution as function of the number of bound electrons for solid-state targets.

magnitude of the shell effects and to compare different targets. An analysis of the peak structures indicates that the shape of the L-shell peak is significantly dependent on Z_t as well as on Z_p. Thus, there are further hidden parameters, neither accounted for in the present fit, nor in the displayed N_b dependence.

Finally, we have also analyzed the widths of the charge-state distributions. In most investigations a width parameter $d = [\sum (q - q_{\text{mean}})^2 P_q]^{1/2}$ (here P_q is the normalized charge-state fraction) is extracted from the experimental data. Partially based on scaling properties worked out in previous work [2,7] we find a reduced width

$$w = d\ Z_p^{-0.27} Z_t^{0.035-0.0009Z_p}\ f(q_{\text{mean}})\ f(Z_p - q_{\text{mean}}), \tag{5}$$

with

$$f(x) = \sqrt{(x + 0.37Z_p^{0.6})/x}. \tag{6}$$

The $Z_p^{-0.27}$ dependence dominates the general trend of the data and the functions f serve to correct for the statistical reduction of the width at either very low or very high mean charge states. Fig. 4 shows the scaled width w as function of N_b. It is seen that most of the scaled solid-state data fall within a narrow band around $w = 0.7$. Again shell effects are clearly visible in this plot. In contrast to Fig. 3, a significant K-shell structure is found for the width. The L-shell maximum of w corresponds to 13 bound electrons. Thus, there is a shift of the structures of $w(N_b)$ with respect to the ones in $q_{\text{mean}}^{\text{experiment}} - q_{\text{mean}}^{\text{fit}}$.

4. Influence on energy-loss calculations

In this section, we present and discuss the influence of the mean projectile charge-state on the energy loss of ions at $v = 14.1v_0$ in amorphous carbon targets. Fig. 5 displays experimental

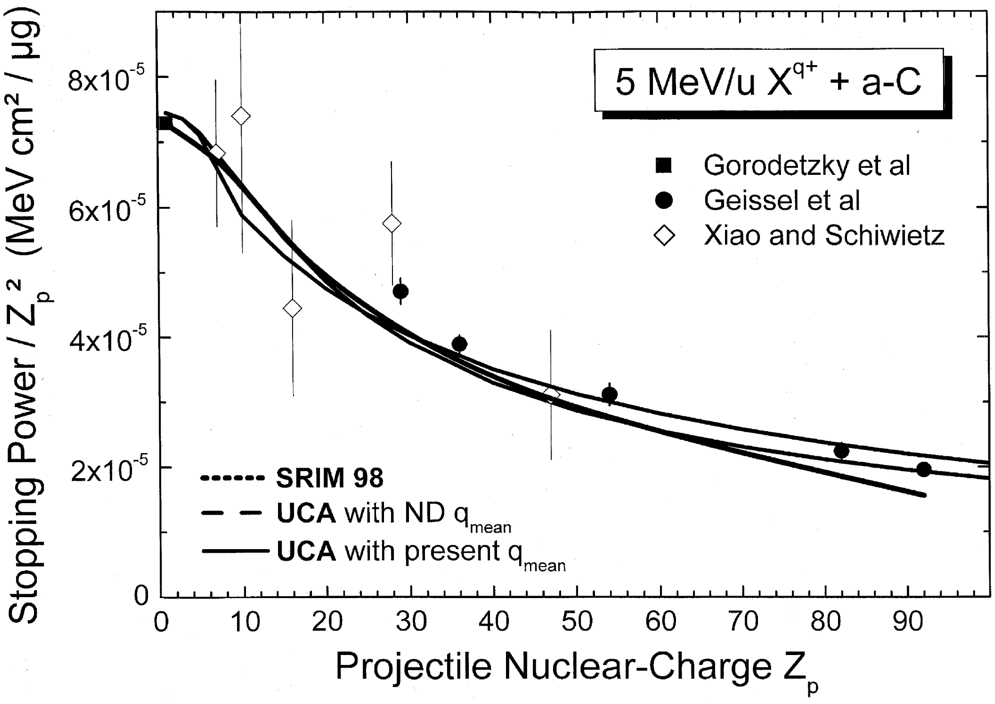

Fig. 5. Energy loss versus projectile nuclear charge for ions at 5 MeV/u in amorphous carbon (see text).

energy-loss results of three groups [14–17] in comparison with predicted energy-loss results. The displayed SRIM-98 results (dotted curve, very similar to TRIM-91) are often taken as a standard for stopping powers [18]. They represent an interpolation of experimental data using multi-parameter fits of measured electronic stopping powers. In contrast, the curves labelled UCA represent results of an ab initio theory (the unitary convolution approximation) [19–21]. This full quantum model accounts for the electronic densities, for the Bloch correction as well as for the projectile screening function. This screening function depends on the projectile charge-state inside the the solid and two different charge-state formulas have been selected as input parameters. The dashed curve labelled ND was calculated with the charge-state formula by Nikolaev and Dmitriev [7] (for $Z_p < 10$ the Bohr stripping criterion as revised by Northcliffe is used). The solid curve was calculated with the present charge-state formula (Eqs. (3) and (4)).

The largest relative deviation between the predictions and the experimental data is 20% for the SRIM-98 results at $Z_p = 92$. This shows that current ab initio results (UCA) can compete even with fits to experimental data. For these fast ions at charge-state equilibrium the influence of electron capture and loss is of minor importance. The most important ingredients that have been neglected in the current UCA treatment are the collective screening and the polarization terms (Barkas effect $\sim Z_p^3$). The first effect yields a reduction and the second an enhancement of the ion-energy loss. The net effect, however, is an increased stopping power. Thus, the UCA results with our present charge-state formula indicate a clear improvement over previous charge-state fits. Note, that the energy-loss code may be downloaded from the Internet. [1]

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[1] The casp code for the UCA and PCA energy-loss theory may be downloaded from http://www.hmi.de/people/schiwietz/casp.html. The next version of the code will include the present charge-state formulas including shell effects.

5. Conclusions

In this work we have investigated mean charge states and widths of charge-state distributions for arbitrary ions in solid and gas targets. The accuracy as well as the range of validity exceeds the one of previous charge-state predictions. We expect that the proposed charge-state formulas can be applied for all heavy positively charged particles (protons, heavy ions as well as muons and other 'elementary' particles) in all gaseous and solid media. For nuclear charges exceeding $Z_p = 2$ they are valid for velocities $v_p > 0.8v_0$. For $Z_p \leqslant 2$ their range of validity is shifted towards higher velocities ($v_p > 2v_0$). He ions in a He gas target, however, constitute a special case where the minimum velocity is $v_p = 2.8v_0$.

For the first time, shell effects in the mean charge state have been extracted on an absolute scale. Until now, however, there is no satisfying theoretical ab initio description of the corresponding structures. The present charge-state predictions (a program with our fit formulas is available[1]) allow to improve stopping-power calculations. Therefore, theoretical uncertainties far below 10% for the energy loss appear to be possible in the near future.

Acknowledgements

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