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Systematics of heavy-ion charge-exchange straggling

P. Sigmund [a,*], A. Schinner [b]

[a] Department of Physics, Chemistry and Pharmacy, University of Southern Denmark, DK-5230 Odense M, Denmark
[b] Department of Experimental Physics, Johannes Kepler University, A-4040 Linz, Austria

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ABSTRACT

The dependence of heavy-ion charge-exchange straggling on the beam energy has been studied theoretically for several ion-target combinations. Our previous work addressed ions up to krypton, while the present study focuses on heavier ions, especially uranium. Particular attention has been paid to a multiple-peak structure which has been predicted theoretically in our previous work.

For high-Z_1 and high-Z_2 systems, exemplified by U in Au, we identify three maxima in the energy dependence of charge-exchange straggling, while the overall magnitude is comparable with that of collisional straggling. Conversely, for U in C, charge-exchange straggling dominates, but only two peaks lie in the energy range where we presently are able to produce credible predictions. For U–Al we find good agreement with experiment in the energy range around the high-energy maximum.

The position of the high-energy peak – which is related to processes in the projectile K shell – is found to scale as Z_1^2, in contrast to the semi-empirical $Z_1^{3/2}$ dependence proposed by Yang et al.

Measurements for heavy ions in heavy targets are suggested in order to reconcile a major discrepancy between the present calculations and the frequently-used formula by Yang et al.

1. Introduction

The phenomenon of charge-exchange straggling was predicted by Flamm and Schumann precisely 100 years ago [1]. In brief, a charged particle penetrating through matter may undergo a sequence of electron capture and loss processes while slowing down. Since the energy loss depends on the ion charge, capture and loss processes give rise to a fluctuation (straggling), in the energy loss. This 'charge-exchange straggling' adds to 'collisional straggling' [2], which also acts in the absence of charge exchange.

On the basis of a theoretical model involving two charge states [3] and a systematic study of hydrogen and helium ions in gas targets [4], the generally accepted view has become that charge-exchange straggling gives rise to a distinct maximum in the dependence of energy-loss straggling on the ion energy for ions with atomic number $Z_1 \geqslant 2$. This view has been strengthened by an analysis of experimental straggling data available in 1991 [5]. That analysis resulted in a frequently-used empirical formula that indicates a straggling peak at an energy $\propto Z_1^{3/2}$ with a peak height $\propto Z_1^{4/3}/Z_2^{1/3}$ relative to the Bohr formula for collisional straggling. Peak heights up to two orders of magnitude above Bohr straggling were predicted for very heavy ions. Experimental support for such pronounced effects came with measurements involving Pb and U ions [6] in the MeV/u energy range. For a recent summary the reader is referred to Ref. [7].

In a joint experimental and theoretical effort [8,9] on krypton and silicon ions in gas targets we confirmed the existence of pronounced maxima, up to two orders of magnitude above the Bohr value. In several cases we found reasonable agreement between experimental data and theoretical predictions, although pronounced discrepancies were found in others. In a parallel theoretical study [10] we predicted that at least two peaks must be found in the energy dependence of charge-exchange straggling. We also found that the leading (high-energy) peak is related to the charge equilibrium between the bare ion and a hydrogen-like ion.

The fact that the leading peak in charge-exchange straggling is related to processes in the K shell of the projectile has a number of implications. Firstly, its position in energy space must be expected to scale as $\propto Z_1^2$ rather than $Z_1^{3/2}$ as proposed in Ref. [5] or, as one might have expected from the Thomas–Fermi model, $\propto Z_1^{4/3}$. Secondly, secondary maxima as well as minima in straggling must be expected likewise to be related to the filling of projectile shells. We wish to address these questions in the present study and to identify ion-target combinations and energy regimes where these effects should be pronounced or, at least, visible in measurements.

The theoretical basis for our work in this area has been a general formalism [11], which expresses charge-exchange straggling by

* Corresponding author.
E-mail address: sigmund@sdu.dk (P. Sigmund).

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transition probabilities or cross sections for all relevant electronic transitions in the projectile. The formalism was first applied in a systematic study of the evolution of energy-loss spectra with the travelled pathlength ([12] and earlier work cited there). Subsequent work focused on straggling in charge equilibrium [13]. Making use of the smooth dependence of the mean energy loss on the ion charge we were able to express charge-exchange straggling by a simple relation involving the evolution of charge fractions with traveled pathlength and the variation of the stopping cross section with the ion charge.

Charge fractions as a function of travelled pathlength are the output of the ETACHA code [14] which, in principle, invokes all those cross sections that are needed in the computation of charge-exchange straggling. Extensive comparisons with experimental data have been performed by Imai et al. ([15] and earlier work cited there), showing qualitative agreement in the general trends.

We use ETACHA output as input into our calculation of charge-exchange straggling. In our previous work with this code [13,10] we had to cope with three limitations: Only ions up to Kr were allowed. The allowed energy range had a lower limit of 1 MeV/u, but the practical lower limit could actually be significantly higher. Moreover, numerical instabilities were frequently found. Specifically, the predicted equilibrium charge state was not always independent of the initial charge state. Since our routine involves small differences between large numbers, it is not easy to identify artifacts introduced by the numerical input.

A revised and expanded edition of the ETACHA code has appeared recently [16]. With an extension of allowed projectiles up to uranium we have now an opportunity to establish scaling relations in Z_1 and Z_2 for both peak position, height and width. At the same time, the relevant energy range expands, since the interesting upper energy limit increases $\propto Z_1^2$, while the lower limit does not. This is relevant for identifying more than one peak in the energy dependence of charge-exchange straggling.

When comparing with experimental straggling data we need to keep in mind that peaks are also present in the energy dependence of collisional straggling [17,18]. Such peaks appear near the stopping maximum and may increase straggling by up to a factor of three above the Bohr value [18]. They are caused by bunching of target electrons and increase in importance with increasing Z_2 where, conversely, charge-exchange straggling decreases in importance.

2. Recapitulation

We report computations on charge-exchange straggling by a procedure developed in Ref. [13] and applied to Kr and Si in gas targets in Refs. [8,10]. Here we briefly summarize the procedure.

The straggling parameter W is defined by

$$W(E,x) = \frac{d}{N dx}\left\langle(\Delta E - \langle\Delta E\rangle)^2\right\rangle, \tag{1}$$

where E denotes the beam energy, ΔE the energy loss of an ion after having traveled a pathlength x, N the number of target atoms per volume and $\langle\ldots\rangle$ an average over many trajectories.

Just as in the case of the mean energy loss, interest is primarily directed towards straggling in a charge-equilibrated beam. According to Refs. [11,13], straggling in charge equilibrium can be written in the form

$$W(E,\infty) \equiv W(E) = \sum_J F_J(E) W_J(E) + W_{\mathrm{chex}}(E), \tag{2}$$

where

$$W_{\mathrm{chex}} = 2N \sum_{JKL} F_J S_{JK} S_L \int_0^\infty dx\,[F_{KL}(x) - F_L], \tag{3}$$

and variables E have been suppressed for clarity. The quantity

$$S_{IJ} = \int T\,d\sigma_{IJ}(T) \tag{4}$$

denotes the stopping cross section for a collision with initial and final states I and J, respectively, and $d\sigma_{IJ}(T)$ the corresponding differential cross section. Moreover,

$$W_J \equiv \sum_L W_{JL}, \tag{5}$$

where

$$W_{JL} = \sum_L \int T^2\,d\sigma_{JL}(T) \tag{6}$$

is the corresponding straggling parameter. If the charge state is I at $x = 0$, $F_{IJ}(x)$ denotes the charge fraction of ions in state J after a path length x. The quantity $F_J \equiv F_J(\infty)$ represents the equilibrium charge fraction.

The first term on the right-hand side of Eq. (2) represents collisional straggling. Eqs. (4) and (5) indicate that energy loss in charge exchange, represented by terms for $I \neq J$, contributes to both collisional and charge-exchange straggling. In the following we consider only the charge-exchange term W_{chex}.

A major simplification was found [13] by making use of the fact that the dependence of the stopping cross section $S_I = \sum_J S_{IJ}$ on the ion charge number q_J can be well approximated by a parabola over a generous interval. For diagonal elements, $I = J$, this assumption was based on calculations by our PASS code [19]. For off-diagonal terms an equivalent behavior was postulated, justified by the fact that energy loss by charge exchange is small compared to collisional energy loss for ions heavier than hydrogen. This point is discussed in appendix A.

For not too light ions, say, $Z_1 \gtrsim 10$, we found that between three neighboring charge states the above parabola can be well approximated by a straight line, so that

$$W_{\mathrm{chex}} \simeq 2N \left(\frac{dS}{dq}\right)_{q=\bar{q}(E)} \left(\frac{dS_{\mathrm{coll}}}{dq}\right)_{q=\bar{q}(E)} G_0(E), \tag{7}$$

where $S \equiv S(q)$ and $S_{\mathrm{coll}} \equiv S_{\mathrm{coll}}(q)$ represent the total frozen-charge stopping cross section and the collisional frozen-charge stopping cross section, respectively, $\bar{q}(E)$ is the mean equilibrium charge at energy E,

$$G_0(E) = \sum_J F_J (q_J - q)\beta_J, \tag{8}$$

and

$$\beta_J = \sum_L q_L \int_0^\infty dx (F_{JL} - F_L). \tag{9}$$

Inspection of Eq. (7) reveals that the effect of charge exchange on W_{chex} is contained in the factor $G_0(E)$, while the factors in front of $G_0(E)$ represent the variation of the stopping cross section with the ion charge. With this, the computational routine involves ETACHA for $G_0(E)$ and PASS for dS/dq.

Although the PASS code distinguishes between S and S_{coll}, this distinction is hardly relevant within the overall accuracy of the theory which, as we shall see, is determined primarily by the ETACHA code. Therefore, following our previous procedure [13,10] we replace Eq. (7) by

$$W_{\mathrm{chex}} \simeq 2N \left(\frac{dS}{dq}\right)^2_{q=\bar{q}(E)} G_0(E), \tag{10}$$

in the following.