Eur. Phys. J. D **23**, 211–215 (2003)
DOI: 10.1140/epjd/e2003-00048-2

THE EUROPEAN
PHYSICAL JOURNAL D

Interplay of charge exchange and projectile excitation in the stopping of swift heavy ions

P. Sigmund[1] and L.G. Glazov[2,1]

[1] Physics Department, University of Southern Denmark, 5230 Odense M, Denmark
[2] Institute of High Current Electronics, Akademichesky 4, 634 055 Tomsk, Russia

Received 23 October 2002 / Received in final form 1st December 2002
Published online 18 February 2003 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2003

Abstract. The electronic energy loss of a dressed ion penetrating through matter is commonly considered as being synonymous with the sum of the excitation energies of the target and the projectile in atomic collisions undergone during the passage. We show that this is not justified in projectile-ionizing collisions and discuss some consequences.

PACS. 34.50.Bw Energy loss and stopping power – 34.50.Fa Electronic excitation and ionization of atoms (including beam-foil excitation and ionization) – 34.70.+e Charge transfer – 52.40.Mj Particle beam interactions in plasmas

1 Introduction

The stopping of swift heavy ions in matter is a complex phenomenon [1] involving several processes that are nonexistent or unimportant in case of light particles such as protons and antiprotons, electrons and positrons. In addition to effects caused by the strong Coulomb interaction between a heavy ion and the target electrons and nuclei, processes affecting the state of the projectile such as charge exchange and projectile excitation need to be considered.

Recent progress has been achieved in the atomistics of target excitation [2,3]. Moreover, a framework for treating statistical aspects of projectile processes has been established some time ago [4] and explored in detail [5,6].

Following up on reference [6] the present note addresses atomistic aspects of charge exchange and projectile excitation, in particular the energetics of projectile ionization where we identified an unclear point in the pioneering paper by Kim and Cheng [7] which seems to have propagated into the literature. Special attention will be paid to the definition of energy loss and its relation to experimental situations.

2 Stopping involving excitation only

For reference we consider an individual collision event between an ion 1 of mass M_1 and velocity $\mathbf{v}_1$ and a target atom 2 at rest. Energy conservation yields

$$\frac{M_1}{2}v_1^2 = \frac{M_1}{2}{v_1'}^2 + \frac{M_2}{2}{v_2'}^2 + W_1 + W_2, \qquad (1)$$

where $\mathbf{v}_1'$ and $\mathbf{v}_2'$ are the velocities of the ion and the target after the collision and W_1 and W_2 the respective excitation energies. The loss of kinetic energy by the projectile is then given by

$$T \equiv \frac{M_1}{2}\left(v_1^2 - {v_1'}^2\right) = W_1 + W_2 + \frac{M_2}{2}{v_2'}^2. \qquad (2)$$

The quantity

$$Q = W_1 + W_2 \qquad (3)$$

is conventionally called the inelastic energy loss.

3 Collisions involving charge exchange

3.1 Definition of energy loss

In the presence of charge exchange we need to define what we mean by energy loss.

If the ion loses an electron during the collision, the change in its translational (center-of-mass) energy is given by

$$\begin{aligned} T' &= \frac{M_1}{2}v_1^2 - \frac{M_1 - m}{2}{v_1'}^2 \\ &= \frac{M_1}{2}\left(v_1^2 - {v_1'}^2\right) + \frac{m}{2}{v_1'}^2 \\ &= \frac{M_1 - m}{2}\left(v_1^2 - {v_1'}^2\right) + \frac{m}{2}v_1^2, \end{aligned} \qquad (4)$$

where m is the electron mass. With $M_1 \gg m$ and, for electronic collisions, $v_1' \simeq v_1$, this reduces to

$$T' \simeq \frac{M_1}{2}\left(v_1^2 - {v_1'}^2\right) + \frac{m}{2}v_1^2 \tag{5}$$

apart from terms that are much smaller.

In particle penetration the point of reference is the *velocity of the nucleus* [5]. Therefore we define the quantity

$$T = \frac{M_1}{2}\left(v_1^2 - {v_1'}^2\right) \tag{6}$$

as the energy loss per collision which, in conjunction with the pertinent cross-section, determines the stopping cross-section. Thus, for a one-electron-loss event we have

$$T = T' - \frac{m}{2}v_1^2. \tag{7}$$

Equation (6) will also be adopted for electron capture.

3.2 Electron loss

For clarity we first consider the case of an ion losing a single electron in a collision.

Energy conservation reads

$$\frac{M_1}{2}v_1^2 = \frac{M_1 - m}{2}{v_1'}^2 + \frac{m}{2}{v_e'}^2 + U_1 + \frac{M_2}{2}{v_2'}^2 + W_1 + W_2, \tag{8}$$

where U_1 is the ionization energy of the ejected electron. The contributions W_1 and W_2 have been kept, except that W_1 now denotes a possible additional excitation (apart from ionization) of the projectile.

From equation (6) we then find

$$T = -\frac{m}{2}{v_1'}^2 + \frac{m}{2}\ {v_e'}^2 + U_1 + W_1 + W_2 + \frac{M_2}{2}{v_2'}^2. \tag{9}$$

With regard to the recoil term $M_2{v_2'}^2/2$ we need to distinguish between nuclear and electronic collisions. In nuclear collisions the momentum transfer is $\leq 2\mu v_1$, where $\mu = M_1M_2/(M_1+M_2)$ is the reduced mass. For sufficiently large momentum transfers $\sim 2\mu v_1$, only the last term in equation (9) is significant. Conversely, in electronic collisions the momentum transfer is $\lesssim 2mv_1$. This makes that term negligible.

The present study addresses electronic processes. Hence we may write

$$T \simeq U_1 + \frac{m}{2}\left({v_e'}^2 - v_1^2\right) + W_1 + W_2, \tag{10}$$

neglecting terms that are much smaller. According to equation (7) we have

$$T' \simeq U_1 + \frac{m}{2}{v_e'}^2 + W_1 + W_2. \tag{11}$$

The above estimate may readily be generalized to a multi-loss event where n_1 electrons are lost from the projectile and n_2 electrons from the target. In an independent-particle notation,

$$T = \sum_{\nu=1}^{n_1}\left[U_{1\nu} + \frac{m}{2}\left({v_{e1\nu}'}^2 - v_1^2\right)\right] + \sum_{\nu=1}^{n_2}\left[U_{2\nu} + \frac{m}{2}{v_{e2\nu}'}^2\right] + W_1 + W_2 \tag{12}$$

and

$$T' = T + n_1\frac{m}{2}v_1^2. \tag{13}$$

3.3 Electron capture

Next, consider a nonradiative event where the ion captures a single electron from a target atom. Energy conservation reads

$$\frac{1}{2}(M_1 - m)v_1^2 = \frac{M_1}{2}{v_1'}^2 + U_2 - U_1' + W_1 + W_2 + \frac{M_2}{2}{v_2'}^2, \tag{14}$$

where U_2 is the binding energy of the captured electron to the target and U_1' its binding energy in the projectile state into which it has been captured.

Leaving out the recoil term again we obtain the energy loss

$$T \simeq \frac{m}{2}{v_1}^2 + U_2 - U_1' + W_1 + W_2 \tag{15}$$

by means of equation (6).

Equation (15) could be generalized to multi-capture events if necessary.

3.4 Inelastic energy loss

We have deliberately avoided the term "inelastic energy loss" Q in the present section because it is somewhat ambiguous. In elementary physics, inelastic energy loss is the difference between the translational energy before and after the collision. This definition reduces equation (8) to $Q = U_1 + W_1 + W_2$. Frequently, however, all electronic energy loss is denoted as inelastic, in which case $m{v_e'}^2/2$ would have to be added. With this Q reduces to T'. Similar considerations apply to the case of capture.

3.5 Capture-loss cycle

For a capture-loss cycle, *i.e.*, a one-electron capture followed by a one-electron loss or *vice versa*, one finds the energy loss

$$T_{\text{cycle}} = T_{\text{loss}} + T_{\text{capture}} = \frac{m}{2}{v_e'}^2 + (U_1 + W_1 + W_2)_{\text{loss}} + (U_2 - U_1' + W_1 + W_2)_{\text{capture}} \tag{16}$$

from equations (10, 15), where the term $mv_1^2/2$ has dropped out. The same result is obtained if the energy loss in capture and loss events is defined by the translational energy T' given by equation (5) for electron loss and its equivalent determined from equation (14) for capture.

3.6 Deposited energy

It is obvious that for composite projectiles a distinction has to be made between the energy lost by the projectile and that deposited in the target [7,8]. In the present notation the latter may be written as

$$D = \frac{m}{2}{v'_e}^2 + W_2 + \frac{M_2}{2}{v'_2}^2 \tag{17}$$

for a collision involving single-electron loss. By means of equation (8) this may be written as

$$D = \frac{M_1}{2}v_1^2 - \frac{M_1 - m}{2}{v'_1}^2 - U_1 - W_1 \tag{18}$$

$$\simeq T + \frac{m}{2}v_1^2 - U_1 - W_1 \tag{19}$$

$$= T' - U_1 - W_1. \tag{20}$$

This assumes energy to be deposited at the collision site. In practical applications it may be necessary to take into account energy transport away from the collision site by secondary electrons ($m{v'_e}^2/2$), Auger electrons (W_2) and recoil atoms ($M_2{v'_2}^2/2$).

4 Reference frames

4.1 Excitation only

The conventional way of treating projectile excitation invokes symmetrization, *i.e.*, interchanging the roles of target and projectile. A simple example is equation (2) where, in the absence of target and projectile ionization, the electronic energy loss

$$T = W_1 + W_2 \tag{21}$$

is simply composed of the excitation energies W_1, W_2 of the projectile and the target.

4.2 Including ionization

Equation (21) has been adopted by Kim and Cheng [7] for *all* excitations, including those into the continuum. It is then tacitly assumed that excitation energies of the projectile are to be measured in a reference frame in which the projectile is at rest. This is unproblematic as long as the excited electron remains bound but becomes questionable when it is ejected.

Consider first excitation and/or ionization of the target described by an energy transfer

$$T_2 = W_2 + \sum_{\nu=1}^{n_2}\left(U_{2\nu} + \frac{m}{2}{v'_{e2\nu}}^2\right). \tag{22}$$

Interchanging the roles of projectile and target implies a transformation to a reference frame moving with the projectile. If velocities in that frame are denoted by

$$\mathbf{u}_{\ldots} = \mathbf{v}_{\ldots} - \mathbf{v}_1, \tag{23}$$

we obtain an energy transfer

$$T_1 = W_1 + \sum_{\nu=1}^{n_1}\left(U_{1\nu} + \frac{m}{2}{u'_{e1\nu}}^2\right). \tag{24}$$

The difference

$$T - T_1 - T_2 = \sum_{\nu=1}^{n_1}\frac{m}{2}\left({v'_{e1\nu}}^2 - {u'_{e1\nu}}^2 - v_1^2\right), \tag{25}$$

T being given by equation (12), is nonvanishing in general. While the last term in the brackets stems from the difference between T and T', the two first terms are of a kinematic nature and cannot be ignored in general. Elimination of $\mathbf{v}'_{e1\nu}$ by means of equation (23) yields

$$T = T_1 + T_2 + \varDelta T \tag{26}$$

with

$$\varDelta T = \mathbf{v}_1 \cdot \sum_{\nu=1}^{n_1} m\mathbf{u}'_{e1\nu}. \tag{27}$$

This reduces to equation (21) in the case of excitation without ionization ($n_1 = 0$) or $\mathbf{u}'_{e1\nu} \to 0$, as it should. Now,

$$\varDelta \boldsymbol{P}_e \equiv \sum_{\nu} m\mathbf{u}'_{e1\nu} \tag{28}$$

is the momentum transfer to ejected projectile electrons. Its component in the direction of $\mathbf{v}_1$ tends to be negative. Hence the expression $T_1 + T_2$ tends to overestimate the energy loss of the projectile.

From momentum conservation and equations (26) and (27) we find

$$T = M_1\mathbf{v}_1 \cdot (\mathbf{v}_1 - \mathbf{v}'_1) = \mathbf{v}_1 \cdot (\varDelta \boldsymbol{P}_2 + \varDelta \boldsymbol{P}_e), \tag{29}$$

where $\varDelta \boldsymbol{P}_2$ is the momentum transferred to the target, and hence

$$T_1 + T_2 = \mathbf{v}_1 \cdot \varDelta \boldsymbol{P}_2, \tag{30}$$

i.e., $T_1 + T_2$ determines the change in velocity of the center-of-mass of the projectile. In case of electron loss, the center-of-mass of what used to be the projectile does not coincide with the position of the nucleus. This is accounted for by the term $\varDelta \boldsymbol{P}_e$.

Thus, the stopping cross-section for events involving electron loss contains a term

$$\varDelta S = \int \varDelta T \, \mathrm{d}\sigma = \mathbf{v}_1 \cdot \int \varDelta \boldsymbol{P}_e \, \mathrm{d}\sigma \tag{31}$$

which is typically negative. Explicit evaluation requires a model for the differential cross-section $\mathrm{d}\sigma$.

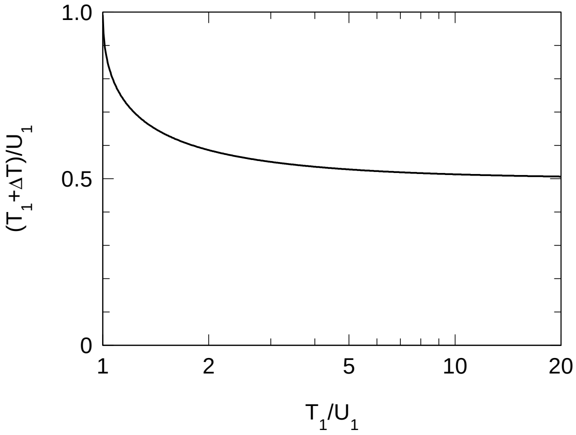

Fig. 1. $T_1 + \Delta T$ *versus* T_1 for binary collisions with an inelastic energy loss U_1. *Cf.* equation (34).

5 An estimate

The correction ΔT must become most pronounced for the highest excitations, where $\mathbf{u}'_{e1\nu}$ is large in magnitude and close to antiparallel with $\mathbf{v}_1$. Following Bohr [9] one may treat such events by binary-collision dynamics, but we need to take into account the ionization energy in the energy balance.

Denoting the momentum transfer from the target atom to a single projectile electron by

$$P = 2mv_1|\cos\varphi|, \tag{32}$$

where φ is the angle between $\boldsymbol{P}$ and $\mathbf{v}_1$, energy conservation leads to

$$T_1 = 2mv_1^2\cos^2\varphi = \frac{m}{2}{u'_e}^2 + U_1 \tag{33}$$

and

$$\Delta T = mv_1 u'_e \cos\varphi = -\sqrt{T_1(T_1 - U_1)}. \tag{34}$$

Figure 1 shows this relation. It is seen that for $T_1 \gg U_1$, *i.e.*, close collisions of the target nucleus with a projectile electron, $T_1 + \Delta T$ approaches the value $U_1/2$ so that T_1 and ΔT nearly cancel each other. From this we find a rough estimate for the contribution from projectile ionization to the stopping cross-section,

$$S_{\text{proj ioniz}} = \int_{T_1 > U_1} (T_1 + \Delta T)\,\mathrm{d}\sigma(T) \sim U_1\sigma_1, \tag{35}$$

where σ_1 is the loss cross-section. This is significantly lower than the value emerging from symmetrization.

6 Discussion

Let us first summarize the problem discussed here in physical terms. As long as all electrons remain in bound states we may consider inelastic energy losses as being superimposed from changes in the electronic configurations of the projectile and the target, respectively,

$$T = T_1 + T_2. \tag{36}$$

If at least one electron leaves the projectile, a choice of reference frame has to be made. In stopping measurements the relevant reference frame is the laboratory system. In that reference frame the kinetic energy of an ejected electron will typically be *lower* than its translational energy before the collision, because in a frame of reference moving along with the projectile, the electron will typically be ejected in the backward direction. Hence, equation (36) ceases to be valid and needs to be replaced by (26).

As far as we are aware this feature has been overlooked in the literature on stopping of partially-stripped ions.

Kim and Cheng [7] performed a pioneering study of the stopping of partially-stripped ions in the Born approximation. Their treatment represents an extension of the Bethe theory [10]. Equation (36) reads $E_{mn} = E_m^{(p)} + E_n^{(t)}$ in their notation, where m and n refer to excited states of the projectile (p) and target (t), respectively. No distinction is made between excitations into bound states and the continuum. As far as we can see the same feature goes again in references [11–14] and subsequent papers by the same groups of authors, all of which address the problem within the Born approximation.

Crawford [8] considered the problem of energy deposition by partially-stripped ions in the stopping medium. While the definition of the quantity studied – which is not strictly equivalent with the one defined in our equation (17) but based on the concept of restricted energy loss – avoids a moving reference frame, the problem returns in the energy balance in Crawford's equation (13) which contains excitation energies $\hbar(\omega_i + \omega_j)$ of the projectile and target, both of which taken in the respective rest frames. Electrons ejected with low velocities in the moving frame have high velocities in the laboratory frame and, therefore, are not included in the restricted-energy-loss function. Conversely, low electron speeds $v'_{1e\nu}$ represent high speeds $u'_{e1\nu}$ which may cause a significant error when included in the energy balance.

Arnau and Echenique [15] considered the stopping of partially-stripped ions in an electron gas. Careful attention was paid to the definition of the energy-loss function[1], but again, target and projectile excitations were treated within the respective rest frames.

Kabachnik and Chumanova [16] studied the impact-parameter dependence of electronic energy loss, including excitation and ionization of the projectile. It is our understanding that the energy and momentum balance in the determination of excitation probabilities has been treated with an adequate degree of rigor. However, their energy-loss function, based on the quantity $\hbar\omega_{fi} = E_i + \epsilon_i - E_f - \epsilon_f$ where $E_{i,f}$ and $\epsilon_{i,f}$ refer to the target and the ion,

[1] Energy loss was defined here as the change in center-of-mass energy T'. According to Section 3.5 this does not lead to ambiguities when only equilibrium stopping is considered.

respectively, does not distinguish between projectile excitation and ionization.

Maynard *et al.* [17] studied projectile excitation on the basis of an average-atom model. In their approximation the velocity of ejected projectile electrons in the moving system, $\mathbf{u}'_{e1\nu}$, was set equal to zero, based on the argument that the excitation probability decreases rapidly with increasing excitation energy. While this approximation can hardly be generally valid, it constitutes an explicit statement that an approximation has been made.

Equation (36) also enters the modified-Bohr stopping theory developed by one of us [18]. Calculations were reported for neutral projectiles in a fixed charge state. According to the discussion in the previous section, estimated stopping cross-sections for projectile ionization must be too high.

A more recent development along that line is the binary theory which, in its original form, addressed target excitation only [2]. Projectile processes were taken into account subsequently [3,19], again in the symmetrized form equation (36).

For oxygen in aluminium, a system studied in great detail, projectile excitation/ionization was found to contribute ~20% of the equilibrium stopping cross-section at 1 keV/u, ~10% at 100 keV/u and less than 1% for at energies exceeding 5 MeV/u. These figures indicate upper bounds on the error made by ignoring ΔT.

Grande and Schiwietz [20] applied their unitary-convolution approximation to the stopping of heavy ions both under equilibrium and non-equilibrium. Projectile excitation and ionization were allowed for, again in the symmetrized form, thus overestimating that contribution.

While inclusion of ΔT is essential in both equilibrium and nonequilibrium stopping, Section 3.5 implies that correct placement of the term $mv_1^2/2$ is of concern mainly in nonequilibrium stopping.

As long as energy loss is defined *via* T, equation (6), this term does not contribute to the energy loss in electron-loss collisions but needs to be included in capture events. This is in accordance with common practice: a captured electron needs to be accelerated to the velocity of the ion [21].

If, alternatively, energy loss is defined over the quantity T', equation (5), the contribution to the stopping cross-section from loss events increases while the one from capture events decreases correspondingly and, in fact, may become negative [22]. While this is not in conflict with conservation laws, it does *not* imply that the projectile gets accelerated.

Comparisons with experimental data on stopping in charge equilibrium – where the number of captures equals that of losses in the average – have occasionally [7,14,23] been performed on the basis of $T_1 + T_2$, ignoring not only ΔT but also the contribution from electron capture to the total stopping cross-section. This implies that the term $mv_1^2/2$ neither enters the contribution from loss nor capture. This may be justified at low velocities if the first term on the right-hand side in equation (15) is negligibly small. However, this velocity range will typically fall outside the range of validity of the Born approximation applied in references [7,14,23].

Apart from the estimate given in Section 5 we have refrained from making numerical evaluations of necessary corrections, because those are specific to the calculational schemes mentioned above. Here we just note that equation (27) has now been incorporated into the PASS code implementing the binary theory [3] and will be taken into account in forthcoming applications.

This work was supported by the Danish Natural Science Research Council (SNF).

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