Multiple electron scattering sequences in C^{q+} + Ar collisions at energies and charge states relevant for carbon ion therapy

K. Tőkési and B. Sulik
Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI), H-4001, Debrecen, Hungary, EU
E-mail: tokesi@atomki.hu

Abstract. Secondary electrons, created in biological tissues by high energy ion impact, may significantly contribute to the fragmentation of small molecules up to single- and double-strand brakes in DNA. Differential spectra of electrons emitted in the collisions of decelerating ions are very important for estimating ion impact radiation damages. In this work, we consider a specific ionization mechanism, the so called Fermi-shuttle acceleration, which can produce fast electrons even by slow ion impact. Focusing our interest to the distal region of the Bragg peak, we calculate electron spectra for singly and doubly charged carbon ions using the classical trajectory Monte Carlo method at 60 keV impact energy, where large fragmentation yields for water molecules have recently been observed experimentally.

1. Introduction
The interaction of an energetic heavy ion with dense matter follows a complex scenario with different types of collision events [1-4]. At a few tens of MeV/u or higher impact energies, the dominant process is ionization for both the projectile ion and the target atoms. Accordingly, the equilibrium charge state of the projectile ion is close to that of the bare projectile nucleus. After decelerated to a few hundred keV/u, excitation and charge transfer processes come into play and the mean charge state of the ions increases. Free electron production by direct ionization or autoionization, however, remains an important contribution to the energy transfer from ions to matter, in the entire energy range during the deceleration of the ions, down to their stopping. For practice, the most interesting impact energy region is where the energy transfer per unit length shows a sharp maximum, usually referred to as the Bragg peak [5]. For carbon ions, e.g., the maximum of the Bragg peak is located at ~300 keV/u, where the mean charge state is q=3.5 [6]. Towards lower energies, the charge state further decreases; e.g., q=1 at 10 keV/u projectile energy. In a recent work [6], it has been shown experimentally that at the low energy side (i.e., in the distal region) of the Bragg peak, the total water fragment ion production had a much flatter profile with projectile energy than would be expected if the water radical formation was assumed to follow the energy-loss profile. This seems to be an important region for understanding the details of the characteristic mechanisms of ion therapy.
A molecule in a biological surrounding can be fragmented in direct ion-molecule collisions or due to the impact of secondary particles (electrons, photons or molecular fragments) emitted in the direct collisions. We note here, that not all the particle-emitting direct collisions lead to a molecular fragmentation, and therefore the yield of secondary processes might be eventually higher than that of the direct fragmentation events. Most of the direct fragmentation processes start with electronic transitions, i.e., the transfer of electrons from the target to the projectile, or with target ionization.
When electrons are removed from the molecule, the repulsive forces between its positive and/or polarized cores may result in a Coulomb explosion [7]. Among the secondary particles emitted in direct collisions, secondary electrons are responsible for many of the further fragmentation events. This has been understood recently, following the focus of interest to radiation damages in biological tissues [8-10]. The majority of the secondary electrons are ballistic electrons ($E \leq 20$ eV). They easily create neutral radicals, ions and electrons. At low energies they may be captured at particular molecular sites or trapped as solvated electrons, contributing to single and double strand breaks in DNA [11]. Even in the sub-excitation energy range (0 –4 eV), resonant processes may enhance molecular dissociation enormously [12].

From the point of view of radiation induced damages, the higher energy part of the secondary electron spectrum is also very important [13,14]. The yield of double-strand breaks induced by electrons in the DNA shows a local maximum at energies of a few hundred eV. These fast electrons have been found to be very effective to segment the DNA molecule [13]. It has also been pointed out, that the fragmentation cross section of water molecules by electron impact maximizes between 50 and 300 eV electron energies [14]. Since free radicals are largely responsible for DNA strand breaks in real tissues, the role of few hundred eV electrons seems to be essential. Accurate knowledge of the amount and distribution of the emitted electrons is especially important in the region of the Bragg peak.

It has been shown recently, that a large amount of unexpectedly fast electrons can be emitted by the impact of slow ions. The responsible mechanism is the so-called Fermi-shuttle acceleration [15-19], where the electron is scattered forward and backward by the incoming heavy projectile ion and the target core before being ejected. Due to the repeated collisions, the electron can be accelerated to relatively high energies.

Up to now, most of the experiments were performed with dressed heavy projectiles and heavy target atoms, where the cross section is large enough for observing such multiple scattering processes. These systems are particularly challenging theoretically to describe the double differential ionization cross sections. So far, no quantum calculations were carried out at such complexity, only classical trajectory Monte-Carlo (CTMC) calculations have been performed for modeling these collisions. Rather surprisingly, for many collision systems quantitative agreement has been found compared with the experimental data [20]. On the other hand, in such a classical calculation, the analysis of the individual particle trajectories allows us to identify the multiple scattering events in a purely classical sense.

In this situation, we decided to perform a combined, systematic experimental and theoretical study of fast electron emission by Fermi-shuttle acceleration in the distal region of the Bragg peak. In the present work, we show the results of the first step in the systematic theoretical studies. We calculated the double differential cross section for electron emission from the collisions of singly and doubly charged carbon ions with argon atoms at 60 keV impact energy (5 keV/u). This energy falls in the distal region of the Bragg peak, where the fragmentation yield for the water molecule has been found to be large [6]. Argon ($Z=18$) is a target which provides a high cross section for the multiple electron scattering process, therefore it is easy to handle experimentally. Moreover, it can be considered as a rather good model for chlorine ($Z=17$) and calcium ($Z=22$) atoms. The latter two atoms are likely to play an important role in producing fast electrons in biological tissues.

2. Calculation

The CTMC simulations have been performed in the three-body approximation. In our CTMC model the three particles are the projectile ion core (P), one atomic active target electron (e), and the remaining target ion core (T). Both projectile and target cores include nucleus and electrons. The three “particles” are characterized by their masses and the interaction potentials between them. For the description of the interaction among the particles, central model potentials (based on Hartree-Fock calculations) have been used. For more details of the CTMC calculations see Ref. [20]. In the calculations, electron emission contributions originating from the target and projectile ionization were calculated separately. The sum of these two components, in the rest frame of the target, is the quantity to be compared with the planned experiments on an absolute scale.
Finally, a random sampling analysis of the individual CTMC trajectories has been performed. In the present stage of the work, only 0.01 percent of the trajectories have been analyzed individually above the energy of 15 eV, so this analysis is still preliminary. At 60 keV total energy (5 keV/u), the maximum energy of the single electron scattering ionization (the so called binary encounter peak) is 10.4 eV, and so 15 eV can be considered as an approximate upper limit of observing projectile-target (P-T) scattering events. The results clearly show that most of the high energy ($E>15eV$) electrons, emitted in these collision systems suffered multiple collisions with the target nucleus and the projectile core.

3. Results

Fig. 1 shows the calculated double differential electron emission spectra. Projectile and target ionization contributions are shown separately, for both singly and doubly charged carbon ions. It is clearly seen that a significant part of the calculated spectra falls above 15 eV. The projectile velocity ($V=0.44$ atomic units, with $\hbar e=m_e=1$) corresponds to 2.6 eV electrons. The binary encounter peak at zero degree is located at $2V$ (10.4 eV). The maximum energy of the triple, projectile-target-projectile ionization contributions can be observed above 15 eV. The binary encounter peak is located at $2V$ (10.4 eV). The maximum energy of the triple, projectile-target-projectile ionization contributions can be observed above 15 eV.

![Figure 1](image.png)

**Figure 1.** Double differential cross section for electron emission as a function of electron energy and observation angle (relative to the beam direction). *(a) and (b): target ionization, (c) and (d): projectile ionization, (a) and (c): singly charged carbon ion projectile, (b) and (d): doubly charged carbon ion projectile.*
(P-T-P) and quadruple (P-T-P-T) scattering at 4V is 41.6 eV, while that for 5-fold and 6-fold sequences it is 93.6 eV (6V). Looking at the figures 1a and 1b (target ionization), it is clear that the calculated cross sections indicate even 10-fold sequences (10V, 2606V), which seems to be unexpected, but such trajectories could really be found in the trajectory analysis. Only future experiments can decide whether this prediction is realistic or not. Similarly, 9-fold scattering sequences are predicted for the projectile ionization (figures 1c and 1d). These strange predictions are all supported by the preliminary random sampling analysis of the CTMC trajectories. We would like to draw attention here to some other features of the calculated spectra. One is that the C+ cross sections are systematically smaller than those for the C+ projectile ions. Moreover, for C+, the low energy part of the projectile ionization cross section is higher than that for the target ionization. A more detailed analysis of these data will be provided, together with those from the presently running other calculations will be provided in a forthcoming publication.

In conclusion, our preliminary results indicate unexpectedly long sequences of multiple scattering in the studied projectile energy range. Since this is already the quasi-molecular collision regime, these classical predictions should be tested by both experiments and quantum calculations. The latter, however, is not very easy. Quantum and classical calculations could be compared first for much simpler collision systems. According to our preliminary statistics, multiple (3-fold or higher order) scattering sequences dominantly contribute (with ~70%) to the electron emission above 15 eV energy. As a next step, the predicted double differential cross sections should be tested by experiment.

Acknowledgement
This work was supported by the Hungarian National Science Foundation, OTKA (Grant Nos: K073703, K72172), the grant "Bolyai" from the Hungarian Academy of Sciences, the TeT Grant No. JP-21/2006, and the Hungarian National Office for Research and Technology.

References
[1] M. E. Rudd and J. H. Macek, Case Studies in Atomic Physics 3 (1976) pp 47-136.
[2] R. A. Baragiola (ed.), Ionization of Solids by Heavy Particles, Nato ASI Series B: Physics, vol. 306 (Plenum Press, New York, 1993)
[3] N. Stolterfoht, R.D. DuBois and R.D. Rivarola, Electron Emission in Heavy Ion-Atom Collisions Springer, Berlin, 1997).
[4] H. Rothard, Nucl. Instr. Meth. B 146, 1 (1998).
[5] Z. Deng, I. Bald, E. Illenberger, M. A. Huels, Phys. Rev. Lett. 95, 153201 (2005).
[6] E. C. Montenegro, M. B. Shah, H. Luna, S. W. J. Scully, A. L. F. de Barros, J. A. Wyer, and J. Lecointre, Phys. Rev. Lett. 99, 213201 (2007)
[7] Z.D. Pesic, J.-Y. Chesnel, R. Hellhammer, B. Sulik, N. Stolterfoht, J. Phys B 37, 1405 (2004). [8] G. Kraft and M. Krämer, Adv. Rad. Biol. 17 (1993) 1.
[9] M. Scholz and G. Kraft, Rad. Prot. Dos. 52 (1994) 29.
[10] Collins G P 2003 Sci. Am. 289(3) 26
[11] Boudaïffa B, Cloutier P, Hunting D, Huels M A and Sanche L 2000 Science 287 1658
[12] Abdoul-Carime H, Gohilke S and Illenberger E, Phys. Rev. Lett. 92 168103
[13] Friedland W, Jacob P, Paretzke H G and Stork T, 1998 Radiat. Res. 150 170
[14] S. W. J. Scully, V. Senthil, J. A. Wyer, E. C. Montenegro, and M. B. Shah, ICPEAC 2005, 24th International Conference on Photonic Electronic and Atomic Collisions Rosario, Argentina, July 20-26, 2005, Abstracts of Contributed Papers, Edited by: F. D. Colavecchia et al., p 324.
[15] S. Suarez, R. O. Barrachina, W. Meckbach, Phys. Rev. Lett. 77 (1996) 474.
[16] U. Bechtold, S. Haagmann, J. Ullrich, B. Bathelt, A. Bohris, R. Moshammer, U. Ramm, C. Bhalla, G. Kraft and H. Schmidt-Böcking, Phys. Rev. Lett. 79 (1997) 2034.
[17] G. Lanzanò, E. De Filippo, D. Mahboub, H. Rothard, S. Aiello, A. Anzalone, S. Cavallaro, A. Elanique, E. Geraci, M. Geraci, F. Giustolisi, A. Pagano, G. Politi, Phys. Rev. Lett. 83 (1999) 4518.
[18] B. Sulik , Cs. Koncz, K. Tökösi, A. Orbán and D. Berényi, Phys. Rev. Lett. 88 (2002) 073201.
[19] B Sulik, N Stolterfoht, R Hellhammer, Z Pesic, Cs Koncz, K Tökösi and D Berényi, Nucl. Inst. and Meth. B 212, 32 (2003).
[20] B. Sulik and K. Tökösi, Adv. Quant. Chem. 52 (2007) 253.