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Energy loss of swift oxygen molecular ions traversing amorphous carbon foils

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Abstract

We have evaluated the energy loss of swift oxygen molecular ions when traversing amorphous carbon foils, considering the velocity dependence of the mean charge of each atomic ion. Our calculations reproduce the main features of the available experimental data and clearly show that the energy loss of molecular ions strongly depends on the molecular axis orientation. © 2000 Published by Elsevier Science B.V.

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1. Introduction

A swift molecular ion bombarding a solid loses its binding electrons in the first atomic layers. The atomic ions resulting from the dissociation of the molecular ion undergo electronic loss and capture processes until they acquire the average charge state corresponding to their velocity. Atomic ions that travel through a solid lose energy mainly through electronic excitations of the stopping medium; however, the energy lost by the atomic ions resulting from the dissociation of a molecular ion is different from the energy lost by the corresponding individual atomic ions moving with the same velocity through the stopping medium. In the former case there is an additional effect resulting from the interaction of

each particle with the electronic excitations created in the solid by its molecular partner, which produces the so called vicinage effects in the energy loss. After the pioneering work by Brandt et al. [1], the interest in the study of the energy loss of molecular ions in solids has not declined and recently many works have been published analysing several related topics [2–9].

The stopping power ratio is a useful magnitude used to measure the vicinage effects in the energy loss of molecular ions. For a homonuclear diatomic molecular ion, the stopping power ratio is defined as

$$R_2 = \frac{S_2}{2S_1}, \quad (1)$$

where S_2 is the stopping power of the target for the molecular fragments (in correlated motion) and S_1 is the stopping power of the target for the atomic constituent, both magnitudes S_1 and S_2 evaluated at the same projectile velocity.

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Tape et al. [10] measured the energy loss of randomly oriented O_2^- ions traversing amorphous carbon foils of several thicknesses, in the energy range $E = 1.6\text{--}2.9$ MeV/atom. They found a stopping power ratio always greater than unity, like the results obtained by Brandt et al. [1] for hydrogen molecular ions. Steuer et al. [11] studied the case of O_2^+ molecular ions incident on amorphous carbon targets, at energies of 1.0 and 1.6 MeV/atom and for several foil thicknesses. Although initially the molecules of the incident beam had the internuclear axis randomly oriented, Steuer et al. [11] measured the energy lost by those fragments leaving the foil at almost zero exit angle between the internuclear axis and the beam velocity, which they denoted as aligned fragments. In contrast to the results obtained for randomly oriented O_2^- ions [10], the stopping power ratio for aligned O_2^+ ions [11] was smaller than unity for the thinner carbon foils, although in both cases the stopping power ratio tends to unity for the thicker foils. The latter effect is because for thicker foils the molecular fragments have longer times to become uncorrelated (mainly due to their mutual Coulomb repulsion). However, even though the former differences could be ascribed to the different orientation of the internuclear axis, to date there is no theoretical treatment that explains satisfactorily these results, only a qualitative discussion was made by Brandt et al. [1,12].

In this work we study the vicinage effects in the energy loss of swift O_2^- and O_2^+ ions traversing a solid foil, paying special attention to their internuclear axis orientation and the velocity dependence of the charge state of each molecular fragment. In Section 2 we present our model, which is based on the dielectric formalism of the stopping power. Our results and a brief discussion about the relationship between the energy loss of a molecular ion and the vicinage effects in the charge state of its atomic ions appear in Section 3. Finally, the main conclusions are discussed in Section 4. Atomic units will be used throughout, except where otherwise stated.

2. Model

The vicinage effects in the energy loss of a diatomic molecular ion depends both on the net charge

of each atomic ion and the internuclear separation between them. The dependence on the velocity of the mean charge of each atomic ion inside the target is considered by evaluating the average number of electrons bound to each atomic ion at a velocity v , namely [13]

$$\langle N \rangle = Z \exp\left(-\frac{0.92 v_r}{Z^{2/3}}\right), \quad (2)$$

where Z is the atomic number of the ion and v_r is the relative velocity of the atomic ion with respect to the valence electrons of the target. Therefore, we have to consider that during its passage through the target, each atomic ion is described by a nucleus dressed with $\langle N \rangle$ electrons. In order to obtain the electronic density associated to these $\langle N \rangle$ electrons we use the general treatment proposed by Brandt and Kitagawa [14]. After the net charge of each ion is obtained, we calculate the energy loss (vicinage effects included) of the molecular fragments as a function of the internuclear separation, which increases with time due to the Coulomb repulsion.

The dielectric formalism [15] is used to calculate the electronic energy loss of an ensemble of atomic ions moving through a medium characterized by a dielectric constant $\epsilon(k, \omega)$, which is a function of the momentum, k , and energy, ω , transferred to the target electrons. The stopping power of a medium for a homonuclear diatomic molecular ion, S_2 , depends on the orientation of the internuclear axis with respect to the beam velocity v . For randomly oriented molecular ions:

$$S_2^{\text{rand}} = 2S_1 + \frac{4}{\pi v^2} \int_0^\infty \frac{dk}{k} [Z - \rho(k)]^2 \frac{\sin(kr)}{kr} \times \int_0^{kv} d\omega \omega \operatorname{Im}\left[\frac{-1}{\epsilon(k, \omega)}\right], \quad (3)$$

and for aligned molecular ions:

$$S_2^{\text{align}} = 2S_1 + \frac{4}{\pi v^2} \int_0^\infty \frac{dk}{k} [Z - \rho(k)]^2 \times \int_0^{kv} d\omega \omega \operatorname{Im}\left[\frac{-1}{\epsilon(k, \omega)}\right] \cos\left(\frac{\omega r}{v}\right), \quad (4)$$

where r is the internuclear distance between the atomic ions, $\operatorname{Im}[-1/\epsilon(k, \omega)]$ is the energy loss

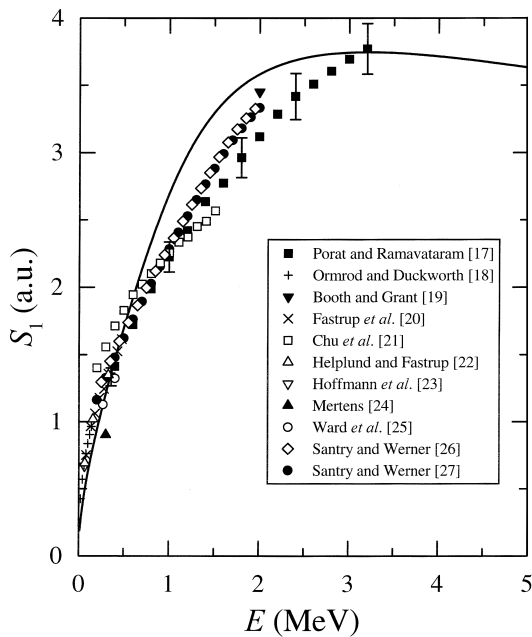


Fig. 1. Stopping power of amorphous carbon for atomic oxygen ions, as a function of the incident energy. Symbols are the experimental data [17–27] and the solid line represents our calculation.

function of the amorphous carbon foil, which is modelled as described in Ref. [16], and $\rho(k)$ is the Fourier transform of the electronic density [14],

$$\rho(k) = \langle N \rangle \left\{ 1 + k^2 \left[\frac{0.23 \langle N \rangle^{4/3}}{(Z - \langle N \rangle / 7)^2} \right] \right\}^{-1}. \quad (5)$$

The stopping power of the target for the case of an individual ion is:

$$S_1 = \frac{2}{\pi v^2} \int_0^\infty \frac{dk}{k} [Z - \rho(k)]^2 \times \int_0^{kv} d\omega \omega \operatorname{Im} \left[\frac{-1}{\epsilon(k, \omega)} \right]. \quad (6)$$

Fig. 1 shows the stopping power of amorphous carbon for an atomic oxygen ion as a function of the projectile energy. Symbols are the experimental data [17–27] and the line represents our calculation using Eq. (6), which agrees reasonably well with the experimental data.

The temporal evolution of the internuclear distance is due to the repulsion between the atomic ions

that constitute the molecular ion. Assuming that the mean charge of each ion only depends on the projectile velocity through Eq. (2), their mutual repulsion is described by the screened Coulomb potential

$$V(r) = \frac{Z - \langle N \rangle}{r} \exp(-r/a), \quad (7)$$

$$a = \begin{cases} v_F / (3^{1/2} \omega_{pl}) & \text{when } v < v_F, \\ v / \omega_{pl} & \text{when } v > v_F, \end{cases}$$

where a is the screening length [29], v_F is the Fermi velocity and ω_{pl} is the plasmon frequency of the target ($v_F = 1.2$ a.u. and $\omega_{pl} = 0.945$ a.u. for amorphous carbon [16]). The effect of the Coulomb repulsion is an increase of the internuclear distance with time. Figs. 2(a) and (b) show the temporal evolution

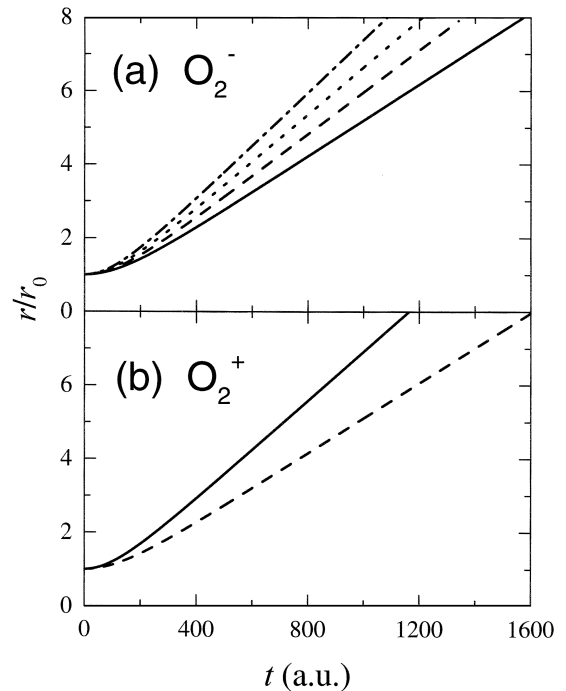


Fig. 2. Normalized internuclear distance, r/r_0 , as a function of the time, t . (a) The energy of the O_2^- ions is 1.6 MeV/atom (—), 2.0 MeV/atom (---), 2.4 MeV/atom (···), and 2.9 MeV/atom (- · - ·). (b) The energy of the O_2^+ ions is 1.0 MeV/atom (—) and 1.6 MeV/atom (---). $r_0(O_2^-) = 2.46$ a.u. and $r_0(O_2^+) = 2.11$ a.u. are the equilibrium internuclear distances [28].

of the normalized internuclear separation, r/r_0 , for O_2^- and O_2^+ ions, respectively (r_0 is the equilibrium internuclear distance: $r_0(O_2^-) = 2.46$ a.u. and $r_0(O_2^+) = 2.11$ a.u. [28]). The energies considered in these figures correspond to the cases for which there are available experimental data for the stopping power ratio of amorphous carbon for oxygen molecular ions: $E = 1.6, 2.0, 2.4,$ and 2.9 MeV/atom for O_2^- [10] and $E = 1.0$ and 1.6 MeV/atom for O_2^+ [11]. It can be seen that the internuclear separation grows faster as the energy increases, because then the screening decreases and the mean charge of the atomic ions increases [13].

3. Results and discussion

According to the previous discussion, Eq. (1) provides the instantaneous stopping power ratio corresponding to a fixed internuclear separation. But as the ions move through the target this separation increases and in order to compare our calculations with the experimental stopping power ratio [10,11], we have to average the instantaneous stopping power ratio over the dwell time, τ ,

$$\langle R_2 \rangle = \frac{1}{\tau} \int_0^\tau dt R_2(r(t)), \quad (8)$$

where we take $\tau = D/v$, being D the foil thickness.

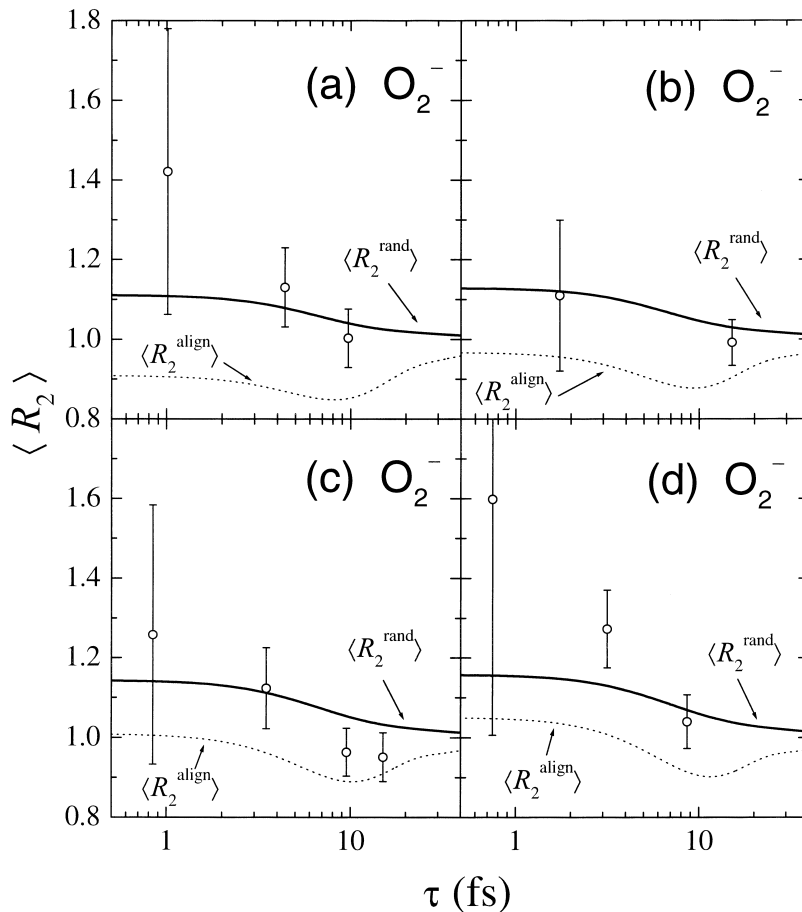


Fig. 3. Stopping power ratio of amorphous carbon, as a function of the dwell time, for randomly oriented O_2^- ions. The energies of the incident O_2^- ions are: (a) $E = 1.6$ MeV/atom, (b) 2.0 MeV/atom, (c) 2.4 MeV/atom, and (d) 2.9 MeV/atom. Circles with error bars are experimental data from Tape et al. [10], solid and thin-dotted lines represent, respectively, our calculations for randomly oriented and for aligned O_2^- ions.

Fig. 3 shows the average stopping power ratio of amorphous carbon for O_2^- ions, as a function of the dwell time and for several incident energies (1.6, 2.0, 2.4, and 2.9 MeV/atom). The symbols (and the corresponding error bars) are the experimental data [10] for randomly oriented O_2^- ions. The solid and thin-dotted lines represent our calculations, respectively, for the cases of randomly oriented and aligned O_2^- ions. It can be seen that $\langle R_2^{\text{rand}} \rangle$ is always greater than one, tending to unity when the dwell time increases. It is important to note that $\langle R_2^{\text{rand}} \rangle$ increases with the projectile energy; this can be understood because the charge of the oxygen fragments is higher for the larger energies [13], resulting in an enhancement of the vicinage effects. The calculated values of $\langle R_2^{\text{rand}} \rangle$ often are inside the error bars and reproduce the same features as the experimental data. On the other hand, the calculated $\langle R_2^{\text{align}} \rangle$ is always less than unity and clearly differs from the experimental data.

In Fig. 4 we present the average stopping power ratio, as a function of the dwell time, corresponding to the atomic ions resulting from the dissociation of O_2^+ when moving through amorphous carbon. The symbols (and the corresponding error bars) are experimental data [11] for aligned O_2^+ ions. Our calculations for this case are depicted by the solid lines and reproduce satisfactorily well the general trends shown by the scarce experimental data; the thin-dotted lines represent our results when the O_2^+ ions are randomly oriented. Even though there is a lack of quantitative agreement, at small dwell times the agreement between experiments and calculations for aligned pairs is better the more energy the projectiles have. It is evident that the calculations corresponding to randomly oriented fragments (thin-dotted line) are always greater than unity and can not reproduce the experimental data. For large dwell times the fragments have time enough to separate, due to Coulomb repulsion, and the vicinage effects tend to disappear ($\langle R_2 \rangle \rightarrow 1$).

It is worth to notice that a small fraction of randomly oriented O_2^+ ions could contribute to the stopping power ratio of the detected aligned fragments. Due to the alignment effect of the wake forces [30,31], that fraction should increase for the larger dwell times and more fragments with an initial random orientation will become oriented with the

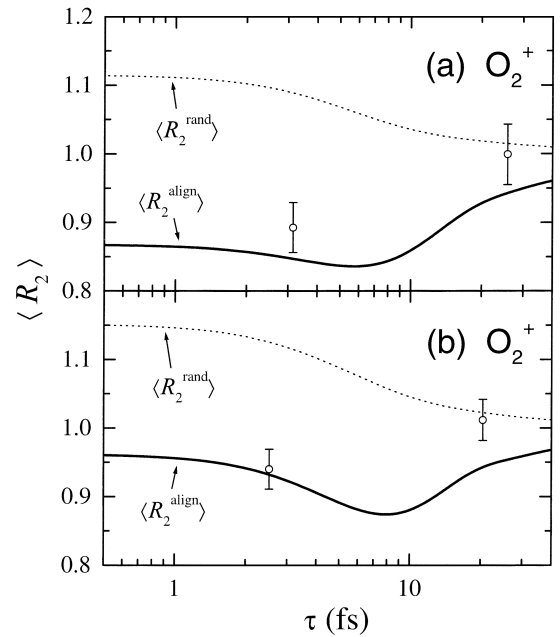


Fig. 4. Stopping power ratio of amorphous carbon, as a function of the dwell time, for aligned O_2^+ ions. The energies of the incident O_2^+ ions are: (a) $E = 1.0$ MeV/atom and (b) 1.6 MeV/atom. Circles with error bars are experimental data from Steuer et al. [11], our calculations are represented by solid lines (for aligned O_2^+ ions) and by thin-dotted lines (for randomly oriented O_2^+ ions).

internuclear axis parallel to the beam velocity. This has a net effect on the stopping power ratio for the larger dwell times, which contains the contribution of both, aligned and random orientations, with the consequence that $\langle R_2^{\text{align}} \rangle$ tends to unity faster, which would improve our present calculations. Analogously (see Fig. 3), a fraction of initially randomly oriented O_2^- ions will be reoriented parallel to the beam velocity due to the alignment effect of the wake forces [30,31]. This will imply the decrease of the average stopping power ratio for the larger dwell times, also improving our calculations.

The present treatment does not consider the vicinage effects in the charge state of the molecular fragments. These effects are due to differences in the charge state of the atomic ions resulting from the fragmentation of the molecular ion and the charge state of their atomic ions considered individually. In general, the charge state of the molecular fragments when they leave the foil is about 20% lower than the

charge state of their individual atomic ions [32,33]. Hence, the implementation of this effect in our calculation of the stopping power ratio would imply the decrease of the vicinage effects in the energy loss (according to the dependence of Eqs. (3) and (4) on the mean charge of the atomic ions), and as a consequence a better agreement with the experimental results for the O_2^+ ions [11] would be expected. Although the implementation of these effects in the case of the O_2^- ions seems to provide a poorer agreement with the experimental data [10], our calculations would still be inside the experimental error bars and reproduce the main trends of the experimental results. However, a recent theoretical calculation [34] suggests that the vicinage effects in the charge state are smaller inside than outside the foil (about 10% inside versus 20% outside the foil). Therefore, in a first approach further changes in the energy loss due to this phenomenon are almost negligible.

4. Conclusions

We have calculated the average stopping power ratio of amorphous carbon for randomly oriented O_2^- and aligned O_2^+ ions, using the dielectric formalism to evaluate the energy loss and considering the Coulomb repulsion of the molecular fragments. In order to improve the calculation of the stopping power ratio we use a realistic description of the energy loss function [16]. Moreover, we have considered that the atomic ions are dressed by electrons, whose average number depends on their velocity [13] and whose spatial distribution is described by means of the Brandt-Kitagawa model [14].

Our calculations show that the average stopping power ratio strongly depends on the molecular axis orientation, being $\langle R_2^{\text{rand}} \rangle > 1$ and $\langle R_2^{\text{align}} \rangle < 1$. These calculations agree satisfactorily well with the experimental results [10,11], explaining their general features. In addition, a qualitative discussion about the alignment effect of the wake forces has been provided, which could help to elucidate the observed discrepancies.

The implementation of the vicinage effects in the charge state of the molecular fragments would improve our calculations, although in a first approach we can neglect them, because they only suppose a

small change in the charge state of the atomic ions (about 10%).

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References

- [1] W. Brandt, A. Ratkowski, R.H. Ritchie, Phys. Rev. Lett. 33 (1974) 1325.
- [2] J.-C. Poizat, J. Remilleux, Phys. Lett. A 34 (1971) 53.
- [3] Y.-N. Wang, T.-C. Ma, Phys. Lett. A 178 (1993) 209.
- [4] C. Tomaschko, D. Brandl, R. Kugler, M. Schurr, H. Voit, Nucl. Instrum. Meth. B 103 (1995) 407.
- [5] C.D. Denton, F.J. Pérez-Pérez, I. Abril, R. Garcia-Molina, N.R. Arista, Europhys. Lett. 35 (1996) 499.
- [6] D. Ben-Hamu, A. Baer, H. Feldman, J. Levin, O. Heber, Z. Amitay, Z. Vager, D. Zajfman, Phys. Rev. A 56 (1997) 4786.
- [7] K. Narumi, K. Nakajima, K. Kimura, M. Mannami, Y. Saitoh, S. Yamamoto, Y. Aoki, H. Naramoto, Nucl. Instrum. Meth. B 135 (1998) 77.
- [8] S. Heredia-Avalos, R. Garcia-Molina, I. Abril, Nucl. Instrum. Meth. B 164–165 (2000) 296.
- [9] N.R. Arista, Nucl. Instrum. Meth. B 164–165 (2000) 108.
- [10] J.W. Tape, W.M. Gibson, J. Remilleux, R. Laubert, H.E. Wegner, Nucl. Instrum. Meth. 132 (1976) 75.
- [11] M.F. Steuer, D.S. Gemmell, E.P. Kanter, E.A. Johnson, B.J. Zabransky, Nucl. Instrum. Meth. 194 (1982) 277.
- [12] W. Brandt, R. Laubert, A. Ratkowski, Nucl. Instrum. Meth. 132 (1976) 57.
- [13] J.F. Ziegler, J.P. Biersack, U. Littmark, The stopping and range of ions in solids, vol. 1 of The Stopping and Ranges of Ions in Solids, Pergamon, New York, 1985.
- [14] W. Brandt, M. Kitagawa, Phys. Rev. B 25 (1982) 5631.
- [15] N.R. Arista, Phys. Rev. B 18 (1978) 1.
- [16] I. Abril, R. Garcia-Molina, C.D. Denton, F.J. Pérez-Pérez, N.R. Arista, Phys. Rev. A 58 (1998) 357.
- [17] D.I. Porat, K. Ramavataram, Proc. Phys. Soc. 77 (1960) 197.
- [18] J.H. Ormrod, H.E. Duckworth, Can. J. Phys. 41 (1963) 1424.
- [19] W. Booth, I.S. Grant, Nucl. Phys. 63 (1965) 481.
- [20] B. Fastrup, P.Hvelplund, C.A. Sautter, K. Dansk, Videnskab. Selskab Mat.-Fys. Medd. 35 No. 10 (1966).
- [21] W.K. Chu, P.D. Bourland, K.H. Wang, D. Powers, Phys. Rev. 175 (1968) 342.
- [22] P. Hvelplund, B. Fastrup, Phys. Rev. 165 (1968) 405.
- [23] I. Hoffmann, E. Jäger, U. Müller-Jahreis, Rad. Effects 31 (1976) 57.

- [24] P. Mertens, *Nucl. Instrum. Meth.* 149 (1978) 149.
- [25] D. Ward, H.R. Andrews, I.R. Mitchell, W.N. Lennard, R.B. Walker, N. Rud, *Can. J. Phys.* 57 (1979) 645.
- [26] D.C. Santry, R.D. Werner, *IEEE Trans. Nucl. Science NS-26* No 1 (1992) 1335.
- [27] D.C. Santry, R.D. Werner, *Nucl. Instrum. Meth. B* 69 (1992) 167.
- [28] K.P. Huber, in: B.H. Billins, H.P.R. Frederikse, D.F. Bleil, R.B. Lindsay, R.K. Cook, J.B. Marion, H.M. Crosswhite, M.W. Zemansky (Eds.), D.E. Gray (coord. Ed.), *American Institute of Physics Handbook*, McGraw-Hill, New York, 1972, p. 7-168.
- [29] W. Brandt, in: S. Datz, B.R. Appleton, C.D. Moak (Eds.), *Atomic Collisions in Solids*, vol. 1, Plenum, New York, 1975, p. 261.
- [30] D.S. Gemmell, *Nucl. Instrum. Meth.* 194 (1982) 255.
- [31] R. Garcia-Molina, I. Abril, C.D. Denton, N.R. Arista, *Phys. Rev. A* 61, June, 2000.
- [32] D. Maor, P.J. Cooney, A. Faibis, E.P. Kanter, W. Koenig, B.J. Zabransky, *Phys. Rev. A* 32 (1985) 105.
- [33] A. Brunelle, S. Della-Negra, J. Depauw, D. Jacquet, Y. Le Beyec, M. Pautrat, *Phys. Rev. A* 59 (1999) 4456.
- [34] S. Heredia-Avalos, R. Garcia-Molina, N.R. Arista, to be published.