Calculations on vicinage effects in the energy loss of fast B_n^+ ($n = 2, 3, 4$) molecules in carbon foils

Santiago Heredia-Avalos [a,*], Rafael Garcia-Molina [a], Isabel Abril [b]

[a] *Departamento de Física, Universidad de Murcia, Apartado 4021, E-30080 Murcia, Spain*
[b] *Departament de Física Aplicada, Universitat d'Alacant, Apartat 99, E-03080 Alacant, Spain*

Abstract

We have evaluated the vicinage effects in the energy loss of the boron fragments resulting from the dissociation of fast B_n^+ ($n = 2, 3, 4$) molecular ions when traversing amorphous carbon foils. We obtain the stopping power ratio as a function of the foil thickness, taking into account the Coulomb explosion among the n fragments of the B_n^+ ion and using the dielectric formalism to describe the electronic excitations produced in the target by the boron fragments. Our results show that the vicinage effects increase with the projectile energy and with the number of molecular constituents, which agree satisfactorily well with the experimental data. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Atomic and molecular beams are currently used as a tool to probe or modify the structure of matter [1]. The study of the differences that appear between the energy loss of a single charge or an ensemble of charges is an interesting topic, which was first analysed both theoretically and experimentally by Brandt et al. [2]; these differences, denominated *vicinage effects*, are caused by the interferences between the electronic excitations produced in the target by each molecular constituent. The stopping properties of the target and the characteristics of the projectile (energy, molecular structure and electronic density) determine these vicinages effects. The stopping power ratio is used to quantify the vicinage effects; it is defined as the energy loss of the molecular ion divided by the energy loss of its atomic constituents considered independently. A stopping power ratio different from unity implies the existence of vicinage effects.

Several works have been devoted to analyse the stopping power ratio for different molecular ions, ranging from the simplest case H_2^+ [2,3] to more complex ones, such as N_2^+ [4], O_2^+ [4] or C_n^+ [5]; and a considerable effort has been made to explain theoretically these results [6–9]. Recently, Narumi et al. [10] have measured the stopping power ratio

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of the boron fragments resulting from the dissociation of fast B_n^+ ($n = 2, 3, 4$) molecular ions incident with energy $E = 0.8$ MeV/atom on amorphous carbon foils. Their results showed that the stopping power ratio increases with the number of molecular constituents, n, and goes to unity for the thicker foils. This general behaviour was justified in a recent work [11]; however, the comparison between the calculations and the experimental results was not very successful. In this paper, we present a calculation of the stopping power ratio as a function of the foil thickness and the projectile structure, which reproduces satisfactorily well the experimental data [10], and we also discuss the dependence of the stopping power ratio on the B_n^+ energy. Atomic units will be used throughout this work, except when otherwise stated.

The model we have used to calculate the stopping power ratio is described in Section 2, and in Section 3 we compare our calculations with the experimental data. Finally a summary is presented in Section 4.

2. Model

When a fast B_n^+ molecular ion bombards a solid target, it loses its binding electrons in the first atomic layers. The n atomic ions resulting from the fragmentation of the B_n^+ molecule undergo a series of electronic capture and loss processes during their travel through the foil. The average number of bound electrons per ion, N, depends on its incident energy, E, as

$$N = Z[1 - q(E)], \tag{1}$$

where Z is the atomic number of the ion and $q(E)$ is its ionization degree, for which we use the parameterization given in [12]. Thereafter, two different processes take place: the interaction between the n atomic ions and the stopping medium and the Coulomb explosion between these ions. These processes can be considered independent due to their different time scales [13]; for this reason we divide our calculation in two parts: (i) we evaluate the stopping power ratio of the n

ions as a function of their instantaneous interatomic distances and (ii) we calculate the temporal evolution of the relative positions of the n atomic ions (due to the Coulomb explosion).

2.1. Vicinage effects

When the n atomic ions move through the foil, vicinage effects appear due to their correlated motion; these are produced by the interference of the electronic excitations induced in the target by each fragment. Therefore, for a given foil, the energy lost by n correlated atomic ions may be different to the energy lost by n independent boron projectiles, depending on the molecular structure and energy. In this paper we will neglect the nuclear elastic collisions with the target atoms, because the high energy of the molecular ions ($E \geqslant 0.4$ MeV/atom). So, we will assume that the projectile energy loss is only due to interactions with the target electrons.

The electronic energy loss of a fast B_n^+ ion moving through a solid can be described by the dielectric formalism [14], where the dynamic properties of the medium are characterized by the dielectric function $\epsilon(k, \omega)$, which depends on the momentum k and the energy ω transferred to the target electrons. This formalism has been satisfactorily applied to calculate the energy loss of hydrogen molecular ions [15] and clusters [16] in solids. The electronic stopping power of a solid for a B_n^+ molecular ion can be written as

$$S_n = nS_1 \left[1 + \frac{2}{n} \sum_{i=1}^{n-1} \sum_{j>i}^{n} I(r_{ij}) \right], \tag{2}$$

where r_{ij} represents the interatomic distance between the i and j atomic ions and $I(r_{ij})$ is the interference function that accounts for the vicinage effects due to the correlation of the atomic ions; S_1 is the stopping power for an isolated B^+ ion, which is calculated with the dielectric formalism as

$$S_1 = \frac{2}{\pi v^2} \int_0^\infty dk \, \frac{\rho^2(k)}{k} \int_0^{kv} d\omega \, \omega \, \text{Im} \left[\frac{-1}{\epsilon(k, \omega)} \right]. \tag{3}$$

$\rho(k)$ is the Fourier transform of the charge density of the atomic ion,

$$\rho(k) = Z - \rho_{e}(k), \tag{4}$$

where $\rho_{e}(k)$ is the Fourier transform of the electronic density of each atomic ion. $\text{Im}[-1/\epsilon(k,\omega)]$ in Eq. (3) is the energy loss function of the amorphous carbon foil, which is modelled by a fit [17] in the optical limit ($k = 0$) through the full excitation energy spectrum, using experimental data [18] for low values of ω and X-ray scattering factors [19] for higher ω. When the B_n^+ ions are randomly oriented with respect to the beam direction, the function $I(r_{ij})$ is given by

$$I(r_{ij}) = \frac{2}{\pi v^2 S_1} \int_0^\infty dk\, \rho^2(k) \frac{\sin(kr_{ij})}{k^2 r_{ij}}$$
$$\times \int_0^{kv} d\omega\, \omega\, \text{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]. \tag{5}$$

We need to specify the electronic density of the atomic ion in order to obtain $\rho(k)$. Instead of the generic orbital proposed in [20] we use hydrogen-like orbitals, which are more suitable to describe the electronic density of low-atomic number ions, as can be seen when comparing both with a more elaborated model [21]. In the range of energy we are interested, the number of electrons per ion is always $N < 4$, so we use 1s and 2s hydrogen-like orbitals, and the electronic density of each boron ion is [22]

$$\rho_{e}(R) = \frac{N\Theta(2-N) + 2\Theta(N-2)}{\pi} Z_S^3 \exp(-2Z_S R)$$
$$+ \frac{(N-2)\Theta(N-2)}{\pi} \left(\frac{Z_S}{2}\right)^3$$
$$\times \left(1 - \frac{Z_S}{2}R\right)^2 \exp(-Z_S R), \tag{6}$$

where $\Theta(\cdots)$ is the Heaviside step function. In the above expression R is the radial distance from the boron nucleus, and Z_S is the effective nuclear charge given by Slater's rules [23]. The Fourier transform of the charge density, Eq. (4), is given by

$$\rho(k) = Z - [N\Theta(2-N) + 2\Theta(N-2)]$$
$$\times \left[1 + \left(\frac{k}{2Z_S}\right)^2\right]^{-2}$$
$$- 2(N-2)\Theta(N-2)\left[\left(\frac{k}{Z_S}\right)^2 - 1\right]$$
$$\times \left[\left(\frac{k}{Z_S}\right)^2 - \frac{1}{2}\right]\left[1 + \left(\frac{k}{2Z_S}\right)^2\right]^{-4}. \tag{7}$$

The stopping power ratio $\mathscr{R}_n$ for the B_n^+ molecular ion was defined as the stopping power S_n for the correlated molecular fragments, divided by n times the stopping power for each isolated fragment, nS_1. Using Eq. (2), $\mathscr{R}_n$ can be written as

$$\mathscr{R}_n = 1 + \frac{2}{n} \sum_{i=1}^{n-1} \sum_{j>i}^{n} I(r_{ij}). \tag{8}$$

The geometries of the B_n^+ ($n = 2, 3, 4$) molecular ions [24] correspond to a linear, an equilateral triangle and a quasi-square structure, respectively. After doing some algebra in the previous equation, we obtain

$$\mathscr{R}_2 = 1 + I(r),$$
$$\mathscr{R}_3 = 1 + 2I(r),$$
$$\mathscr{R}_4 = 1 + 2I(r) + I(\sqrt{2}r), \tag{9}$$

with r being the interatomic distance between the nearest atomic neighbours in each molecular ion.

In Fig. 1 we present the interference function, $I(r)$, for three energies of the B_n^+ projectile ($E = 0.4$, 0.8 and 1.2 MeV/atom). A positive (negative) value of the interference function means an increment (diminution) of the energy lost by the correlated boron fragments with respect to the same number of noninteracting atomic ions. It is worth to notice that when the energy increases: (i) the position of the minimum value of $I(r)$ shifts towards larger distances r, (ii) the absolute value of the minimum $I(r)$ decreases and (iii) the region of interatomic distances where $I(r) < 0$ widens.

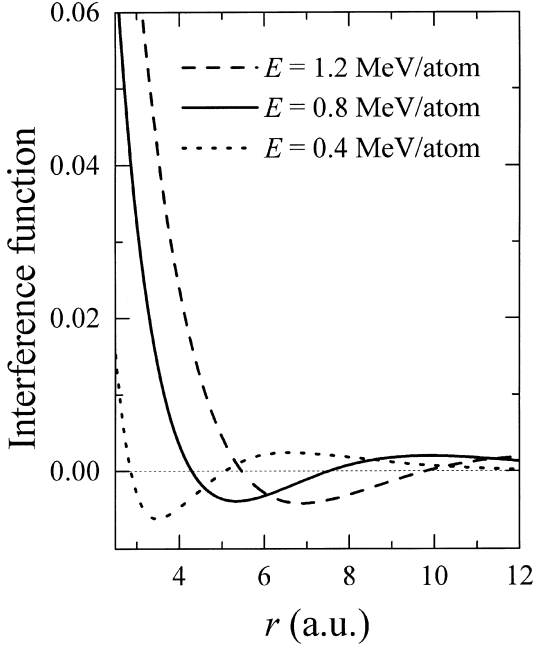To compare our calculations with the experimental data, we average the stopping power ratio, Eqs. (9), over the dwell time τ,

Fig. 1. Interference function versus interatomic distance for three energies of the B_n^+ molecular ion: $E = 0.4$ MeV/atom $(\cdots)$, $E = 0.8$ MeV/atom (—) and $E = 1.2$ MeV/atom (– –).

$$\langle \mathscr{R}_n \rangle = \frac{1}{\tau} \int_0^\tau \mathrm{d}t \, \mathscr{R}_n[r(t)], \tag{10}$$

where $r(t)$ is the temporal evolution of the inter-atomic distance, due to the Coulomb explosion, and $\tau = D/v$, with D being the foil thickness and v the B_n^+ initial velocity.

2.2. Coulomb explosion

The Coulomb explosion between the n atomic ions is governed by a screened Coulomb potential; for an atomic ion with charge $Z - N$ the potential it creates at a distance R is given by

$$V(R) = \frac{Z - N}{R} \exp{(-R/a)}, \tag{11}$$

where a is the screening length ($a = v/\omega_{pl}$ if $v > v_F$ and $a = v_F/(3^{1/2}\omega_{pl})$ otherwise [25]), v_F the Fermi velocity and ω_{pl} is the plasmon frequency of the target electrons. For amorphous carbon we use the value $\omega_{pl} = 0.945$ a.u., which gives the main contribution to the energy loss spectrum [18].

In order to find the temporal evolution of the interatomic distances between the fragment ions, it is necessary to specify the initial structure of the boron molecular ion, for which we use the B_n^+ equilibrium structures given in [24]. The inter-atomic distance of the linear B_2^+ ion is $r_2 = 4.164$ a.u. The structure of the B_3^+ ion is an equilateral triangle with side length $r_3 = 2.930$ a.u. The B_4^+ ion has a quasi-square structure, with the angles be-tween its constituents being alternately 89.92° and 90.08° and whose side length always is $r_4 = 2.913$ a.u.; for the sake of simplicity, in what follows, we will take B_4^+ as a perfect square.

Due to the symmetry of the initial structures of the B_n^+ ($n = 2, 3, 4$) ions, the Coulomb explosion changes only their size preserving their initial shape. Then, one single variable is enough to characterize the temporal evolution of each mo-lecular structure. We take this variable to be the interatomic distance between the nearest atomic neighbours, $r(t)$, which is obtained solving nu-merically the Newton equation for the temporal evolution of the n atomic ions subjected to their mutual Coulomb repulsion. Fig. 2 shows how the interatomic distances for B_2^+, B_3^+ and B_4^+ change with time, when the projectile energy is 0.8 MeV/atom; note that $r(t)$ is normalized to each initial distance, r_n, which is different for each boron molecular ion. We also depict $r(t)$ for B_2^+ when $E = 0.4$ MeV/atom and $E = 1.2$ MeV/atom; it can be observed that $r(t)$ grows faster when the B_2^+ energy increases, which is due to the behaviour of both the ionization degree (increasing with E) and the screening length (increasing with E). The Coulomb repulsion is maximum for the B_3^+ mo-lecular ion, because the net Coulomb force acting on each atomic constituent depends on the initial molecular structure.

3. Results and discussion

In Fig. 3 we show the average stopping power ratio for B_2^+, B_3^+ and B_4^+ ions moving through amorphous carbon, as a function of the foil thickness D. The symbols (and error bars) corre-spond to experimental data [10] for the case of 0.8 MeV/atom incident energy. Our calculations for
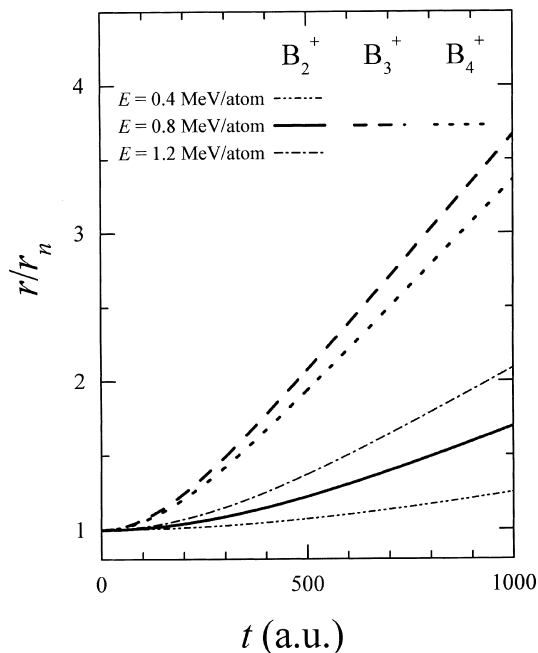
Fig. 2. Evolution of the interatomic distance with time for B_n^+ ($n = 2, 3, 4$) molecular ions with $E = 0.8$ MeV/atom: B_2^+ (—), B_3^+ (– –) and B_4^+ (· · ·). The initial interatomic distances for each molecular ions are: $r_2 = 4.164$ a.u. for B_2^+, $r_3 = 2.930$ a.u. for B_3^+ and $r_4 = 2.913$ a.u. for B_4^+. The temporal evolution of the interatomic distance for B_2^+ is also depicted when the incident energy is $E = 0.4$ MeV/atom (thin – · · –) and $E = 1.2$ MeV/atom (thin – · –).

Fig. 3. Average stopping power ratio for B_2^+, B_3^+ and B_4^+, as a function of the carbon foil thickness, D. Dotted, solid and dashed lines correspond to $E = 0.4$ MeV/atom, $E = 0.8$ MeV/atom and $E = 1.2$ MeV/atom, respectively. The experimental data [10] correspond to $E = 0.8$ MeV/atom.

several B_n^+ energies are represented by lines: 0.4 MeV/atom (dotted line), 0.8 MeV/atom (solid line) and 1.2 MeV/atom (dashed line). According to other works by the Kyoto group [26], a density of 1.7 g/cm^3 for the amorphous carbon foils has been used in our calculations.

In the three cases depicted in Fig. 3 $\langle \mathscr{R}_n \rangle$ decreases the thicker is the foil, tending to unity as the target thickness increases. This is a general trend, because $r(t)$ grows with the dwell time, and the vicinage effects are smaller the larger is the interatomic distance between the atomic ions.

The average stopping power ratio increases with E, for the three boron molecular ions analysed. This behaviour can be understood in the light of Fig. 1, where it can be seen that for values of r of the order of the initial interatomic distances, $I(r)$ is higher when the energy increases.
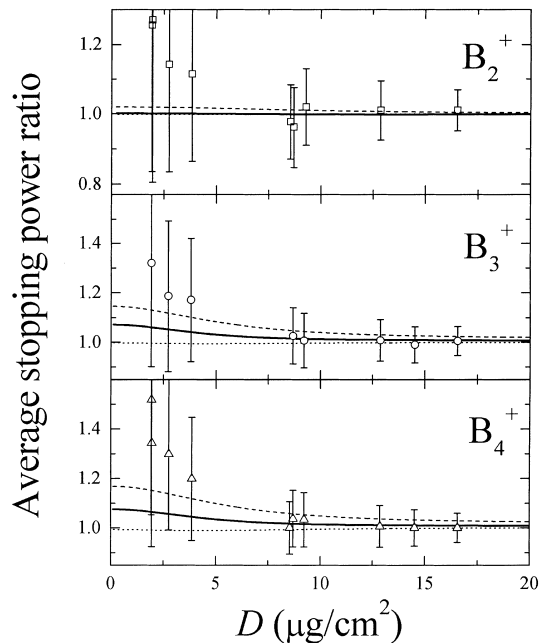
The average stopping power ratio is negative for the thinner foils when $E = 0.4$ MeV/atom, because most of the dwell time the interatomic distance is within the range where $I(r) < 0$, as shown in Fig. 1. For the other energies ($E = 0.8$ and 1.2 MeV/atom) the negative part of the interference function begins to develop later (for larger values of r), and most of the contribution to $\langle \mathscr{R}_n \rangle$ comes form positive values of $I(r)$.

It is interesting to notice that $\langle \mathscr{R}_2 \rangle$ is very close to unity independently of the projectile energy, which is due to the large value of the initial interatomic distance between the two atoms of the B_2^+ molecular ion. This reveals the importance that the initial structure of the molecular ion has in the vicinage effects.

In Fig. 3 we observe that the average stopping power ratio increases with the number of atoms in the boron molecular ion, B_n^+. This is so because the initial distance r_n between the boron atoms

decreases from $n = 2$ to 4, and also because the interferences grow with the number of neighbours, cf. Eqs. (9). The most important vicinage effects appear for the case of B_4^+, due to the number of neighbours, to their initial structure and also because the Coulomb repulsion is smaller than for the B_3^+ ion (see Fig. 2).

4. Summary

The average stopping power ratio of the atomic ions dissociated from fast B_n^+ ($n = 2, 3, 4$) molecular ions moving through amorphous carbon foils has been calculated as a function of the foil thickness and for several projectile energies. The calculation incorporates the Coulomb explosion between the fragment ions and uses the dielectric formalism to evaluate the energy loss and vicinage effects of the ions. The behaviour of the average stopping power ratio shows an increase with the projectile energy and with the size of the boron molecular ion.

Comparison of our calculations with recent experimental data by Narumi et al. [10] for B_n^+ ($n = 2, 3, 4$) molecular ions incident with $E = 0.8$ MeV/atom in amorphous carbon foils is fairly good. The general trends of the experiments are well reproduced by our results, which are within the error bars for the thinner foils and show a good agreement for the thicker foils, where the experimental uncertainty is smaller.

Acknowledgements

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