Transient charge-state effect in the energy loss of swift molecular ions in solids

Rafael Garcia-Molina and Santiago Heredia-Avalos
Departamento de Física, Universidad de Murcia, Apartado 4021, E-30080 Murcia, Spain
(Received 20 June 2000; published 16 March 2001)

The energy loss of the fragments resulting from the dissociation of fast N_2^+ molecular ions when traversing amorphous carbon foils has been calculated. In addition to using the dielectric formalism to describe the electronic excitations of the stopping medium and the Coulomb repulsion to account for the variation of internuclear distances, we have also taken into account the transient charge states of each fragment. The inclusion of this effect was crucial in order to obtain a satisfactory comparison of our calculations with the available experimental data.

DOI: 10.1103/PhysRevA.63.044901 PACS number(s): 34.50.Bw, 36.40.−c

When a swift charged particle moves through a solid it loses energy by exciting the electrons of the stopping medium; this phenomenon is known as electronic energy loss. But when two charged particles move in a correlated way through a solid, the energy they lose is not the sum of the energies lost by each one of the (otherwise individual) particles. In the latter case there are additional effects, such as that resulting from the interaction of each particle with the electronic excitations created in the solid by its molecular partner, which produces so called vicinage or interference effects. Brandt *et al.* [1] showed, both experimentally and theoretically, that there was a difference between the energy loss of the fragments of H_2^+ and H_3^+ molecular ions and the energy loss of individual protons. After that work, many papers appeared discussing several topics related to the energy loss of swift molecules and clusters through solids. Recent reviews [2,3] present the state of the art on this subject to date, analyzing the rich phenomenology that appears in this type of interaction.

After such intensive study in this field, there is still no proper explanation of some experimental results. One of these experiments corresponds to the measurement [4] of the energy loss of N_2^+ molecular ions with their internuclear axis aligned parallel to the beam direction. Although several works [2,5–11] have attempted to explain these experimental data in terms of the interferences of the wake potentials induced in the medium by projectiles moving in a correlated manner, none provided a satisfactory agreement with the experimental data in the whole range of projectile energies and foil thicknesses.

In this Brief Report we incorporate an additional effect in the calculation of vicinage effects in the energy loss of molecular ions. We have taken into account the finite time needed by the molecular fragments to reach their equilibrium charge states when moving through the solid [12]. This transient charge-state effect will prove to be crucial in providing a satisfactory agreement with the experimental results [4].

The measurements by Maor *et al.* [13] of the average charge of N_n^+ $(n=1,2)$ ions, with an energy of 2 MeV/atom, after traversing thin carbon foils showed that the average charge of these ions was not constant in the first atomic layers; the nitrogen atomic ions had to cross several atomic layers before they acquired their equilibrium average charge. These results suggest the need to consider the transient charge states of atomic ions in a calculation of the vicinage effects in energy loss, being particularly important as the target foil becomes thinner.

In what follows we explain the procedure we have followed to evaluate the stopping power ratio corresponding to the experiments reported by Steuer *et al.* [4], although the procedure can easily be extended to other situations. Our treatment is divided into two steps. In the first one we obtain the vicinage effects in the energy loss of the molecular ions, which are evaluated for the instantaneous internuclear separation. The screened Coulomb repulsion between the molecular fragments, which produces an increase of their internuclear separation during the dwell time, is properly taken into account in the second step. In what follows we will work in atomic units, except where otherwise stated.

In order to include the transient charge-state effect in our calculations we will evaluate the average number of electrons surrounding each atomic ion as a function of the time elapsed after it enters the solid, $N(t)$. The calculations are simpler if we neglect the vicinage effects in the charge state of the molecular fragments. Therefore, we will suppose that the transient charge states are identical for atomic and molecular fragments of the same nature. For a projectile with velocity v, we obtain $N(t)$ by means of

$$N(t)=N_\infty-(N_\infty-N_0)\exp(-t/\tau), \tag{1}$$

where N_0 and N_∞ are the average number of electrons just at the entrance of the foil or at equilibrium in the steady state inside the foil, respectively. We will assume $N_0=Z-0.5$ because the nitrogen atomic ions were dissociated from N_2^+ ions, and N_∞ was taken from Ref. [14]. τ is the ionization time, which can be estimated as $\tau=(nv\sigma)^{-1}$, n being the number of target valence electrons per unit volume ($n=4\times10^{23}$ cm^{-3} for amorphous carbon) and σ the ionization cross section, for which we take the approximate value $\sigma=10^{-17}$ cm^2 [15]. This estimation of the ionization time is in good agreement with the result obtained by fitting with Eq. (1) the average transient charge of N^+ ions obtained by Maor *et al.* [13].

The stopping power ratio is useful to quantify the vicinage effects in the energy loss of molecular ions. It is defined as the energy loss of the molecular fragments (in correlated motion) divided by the sum of the energy lost by each one of

 ©2001 The American Physical Society

these fragments (considered individually), all the projectiles at the same velocity and having traversed the same foil thickness. In the case of a N_2^+ molecular ion, this is

$$R_2=\frac{\Delta E(N_2^+)}{2\Delta E(N^+)}=\frac{S_p(N_2^+)}{2S_p(N^+)}. \tag{2}$$

In the above expression we have used the relation between the stopping power S_p and the energy loss ΔE and foil thickness D, $S_p=\Delta E/D$.

The energy loss and the vicinage effects are due to electronic excitations induced in the medium by the passage of the projectile, and they are calculated in the framework of the dielectric formalism [16]. For a given internuclear separation r, the stopping power ratio corresponding to the fragments of a N_2^+ ion that travel aligned with the incident velocity v is written as

$$R_2(r)=1+I_{\text{align}}(r). \tag{3}$$

The value of the interference function $I_{\text{align}}(r)$ is

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

where S_p is the stopping power of the target for an atomic ion moving with velocity v,

$$S_p=\frac{2}{\pi v^2}\int_0^\infty dk\frac{[Z-\rho(k)]^2}{k}\int_0^{kv} d\omega\,\omega\,\text{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right], \tag{5}$$

and Z is the atomic number of the ion. The energy loss function $\text{Im}[-1/\epsilon(k,\omega)]$ is a property of the stopping medium, and it gives the probability of producing an electronic excitation with energy ω and momentum k. We will use the energy loss function model described in Ref. [17]. The Fourier transform $\rho(k)$ of the electronic density corresponding to each atomic ion is obtained from the Brandt-Kitagawa model [18]:

$$\rho(k)=N(t)\left\{1+k^2\left[\frac{0.23N(t)^{4/3}}{[Z-N(t)/7]^2}\right]\right\}^{-1}. \tag{6}$$

Notice how $\rho(k)$ varies on time through its dependence on the average number of electrons $N(t)$.

According to Eq. (4), we have to calculate the stopping power in order to evaluate the interference function. In Fig. 1 we present the stopping power of amorphous carbon for N^+ ions as a function of their energy, when the average charge is at equilibrium, i.e., $N(t)=N_\infty$. Symbols are the experimental data [19–30] and the solid line is our calculation [using Eqs. (5) and (6)]. The agreement of our results and the experimental data is reasonably good.

The Coulomb explosion between the atomic ions resulting from the dissociation of the N_2^+ ions is taken into account

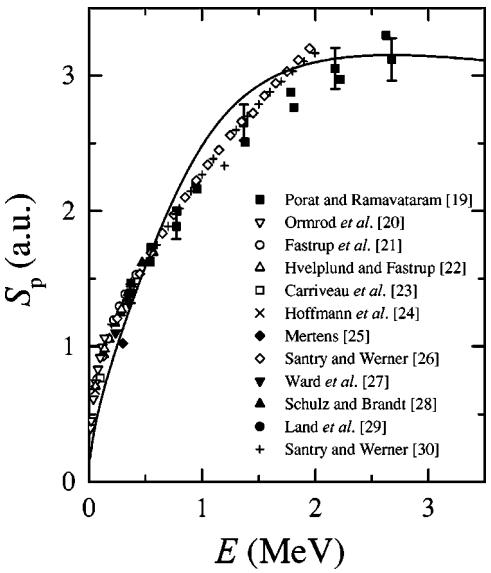

FIG. 1. Stopping power of amorphous carbon for N^+ ions, as a function of the incident energy. The symbols represent the experimental data [19–30] and the solid line corresponds to our calculations using Eqs. (5) and (6).

by numerically solving the temporal evolution of the internuclear distance $r(t)$. Because the interaction between the nitrogen atomic ions is screened by the target electrons, we use the screened Coulomb potential

$$V(r)=\frac{Z-N(t)}{r(t)}\exp\left(\frac{-r(t)}{a}\right), \tag{7}$$

where $a=v_F/(3^{1/2}\omega_{pl})$ when $v<v_F$, and $a=v/\omega_{pl}$ when $v>v_F$; ω_{pl} is the plasmon energy and v_F is the Fermi velocity of the target electrons. For amorphous carbon we use $\omega_{pl}=0.86$ a.u. and $v_F=1.2$ a.u. The effect of the Coulomb explosion is an increase of the internuclear distance with time. In order to find $r(t)$ it is necessary to specify the value of the initial internuclear distance of the nitrogen molecular ion, r_0, for which we use $r_0=2.11$ a.u. [31].

To compare our theoretical results with the experimental data we average over the dwell time D/v the stopping power ratio evaluated for each internuclear separation

$$\langle R_2\rangle=\frac{v}{D}\int_0^{D/v} dt R_2[r(t)]. \tag{8}$$

The goal of this paper is to use the transient charge states of the projectile, Eq. (1), in the calculation of the energy loss of swift molecular ions; it is worth noting that the interference function, the stopping power, and the Coulomb explosion depend on the time not only by means of the internuclear distance, but also through the transient charge states of the atomic ions. Consideration of the latter effect produces a significant improvement in our calculations of the stopping power ratio, as can be seen in Fig. 2, where we show the average stopping power ratio corresponding to the aligned

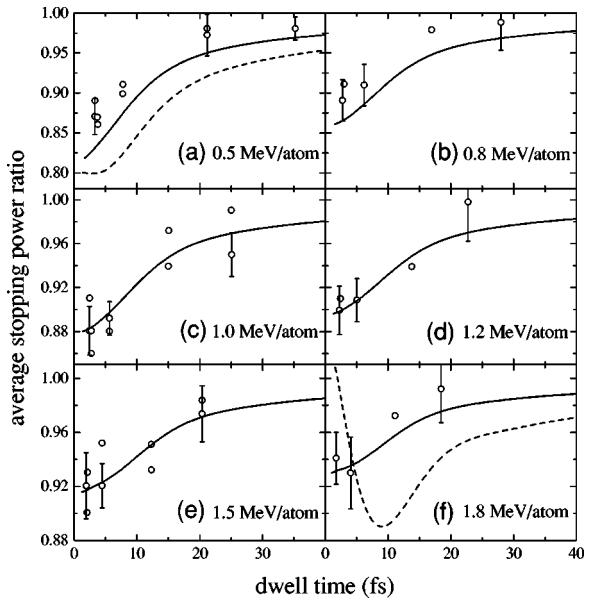

FIG. 2. Average stopping power ratio of amorphous carbon for the fragments of N_2^+, as a function of the dwell time. The experimental data (symbols and error bars) are from Steuer *et al.* [4]. The solid and dashed lines represent our calculations with and without the transient charge-state effect, respectively. The energies of the incident N_2^+ ions are $E=$ (a) 0.5 MeV/atom, (b) 0.8 MeV/atom, (c) 1.0 MeV/atom, (d) 1.2 MeV/atom, (e) 1.5 MeV/atom, and (f) 1.8 MeV/atom.

fragments resulting from the dissociation of N_2^+ when moving through amorphous carbon, as a function of the dwell time. The symbols (and the corresponding error bars) are experimental data [4] for the projectile energy range 0.5–1.8 MeV/atom; the solid lines represent our calculations for aligned pairs when the transient charge states of the nitrogen atomic ions are taken into consideration. For comparison purposes, Figs. 2(a) and 2(f) depict the corresponding calculations without consideration of these transient charge-state effects. It can be seen that the latter calculation reproduces the general trends of the experimental data only at the lowest energy, but fails at the highest energy. The former calculations of the stopping power ratio agree remarkably well with the experimental results: $\langle R_2 \rangle$ is always smaller than unity, going to unity for the larger dwell times (due to the Coulomb repulsion), and the vicinage effects tend to disappear as the projectile energy increases.

It is important to note that a small fraction of randomly oriented N_2^+ ions could contribute to the stopping power ratio of the detected aligned fragments. This is due to the alignment effect of the wake forces [31,32], which is more significant for the larger dwell times; therefore, some fragments with an initially random orientation will be detected with the 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 contributions of both aligned and random orientations, with the consequence that the calculated $\langle R_2 \rangle$ will tend to unity faster; which would improve our present calculations.

In conclusion, we have calculated the average stopping power ratio of amorphous carbon foils for swift aligned N_2^+ ions, in the framework of the dielectric formalism and taking into account the Coulomb explosion between the molecular fragments. We have included in our calculations the evolution of the charge states of each atomic ion, from its initial average charge (when it enters the foil) until it acquires its equilibrium average charge. Although our model was restricted to first order perturbation theory, the inclusion of higher order terms would not contribute sensibly to the stopping power ratio [33]; on the other hand, the neglect of nuclear scattering is reasonable because the contribution to the energy loss is small in the velocity range being discussed and, in addition, the aligned fragments detected are those that have not suffered a significant nuclear scattering; finally, the importance of inhomogeneities in the target thickness has not been discussed because these data were not provided in the original experiments. The comparison with experimental data [4] is better by far than other attempts [2,5–11] to explain these experiments theoretically. This proves that the transient charge states of the projectiles have an important effect in evaluation of the average stopping power ratio, especially for shorter dwell times and higher energies.

This work was supported by the Spanish DGEC through Project No. PB96-1118 and by the Spanish MEC through a research grant to S.H.A.

[1] W. Brandt, A. Ratkowski, and R. H. Ritchie, Phys. Rev. Lett. **33**, 1325 (1974).
[2] J. Jensen and P. Sigmund, Phys. Rev. A **61**, 032903 (2000).
[3] N. R. Arista, Nucl. Instrum. Methods Phys. Res. B **164-165**, 108 (2000).
[4] M. F. Steuer, D. S. Gemmell, E. P. Kanter, E. A. Johnson, and B. J. Zabransky, Nucl. Instrum. Methods Phys. Res. **194**, 277 (1982).
[5] M. F. Steuer, D. S. Gemmell, E. P. Kanter, E. A. Johnson, and B. J. Zabransky, IEEE Trans. Nucl. Sci. **NS-30**, 1069 (1983).
[6] J. Kemmler, P. Koschar, M. Burkhard, and K. O. Groeneveld, Nucl. Instrum. Methods Phys. Res. B **12**, 62 (1985).
[7] M. F. Steuer, Nucl. Instrum. Methods Phys. Res. B **13**, 137 (1986).
[8] M. F. Steuer and R. H. Ritchie, Nucl. Instrum. Methods Phys. Res. B **33**, 170 (1988).
[9] M. F. Steuer and R. H. Ritchie, Nucl. Instrum. Methods Phys. Res. B **40/41**, 372 (1989).
[10] Y.-N. Wang and T.-C. Ma, Phys. Lett. A **178**, 209 (1993).
[11] T. Takamoto and T. Kaneko, Nucl. Instrum. Methods Phys. Res. B **153**, 21 (1999).
[12] T. Schenkel, M. A. Briere, A. V. Barnes, A. V. Hamza, K. Bethge, H. Schmidt-Böcking, and D. H. Schneider, Phys. Rev. Lett. **79**, 2030 (1997).
[13] D. Maor, P. J. Cooney, A. Faibis, E. P. Kanter, W. Koenig, and B. J. Zabransky, Phys. Rev. A **32**, 105 (1985).

[14] J. F. Ziegler, J. P. Biersack, and U. Littmark, *The Stopping and Range of Ions in Solids* (Pergamon, New York, 1985).
[15] N. Bohr, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. **18**, 8 (1948).
[16] N. R. Arista, Phys. Rev. B **18**, 1 (1978).
[17] I. Abril, R. Garcia-Molina, C. D. Denton, F. J. Pérez-Pérez, and N. R. Arista, Phys. Rev. A **58**, 357 (1998).
[18] W. Brandt and M. Kitagawa, Phys. Rev. B **25**, 5631 (1982).
[19] D. I. Porat and K. Ramavataram, Proc. Phys. Soc. London **78**, 1135 (1961).
[20] J. H. Ormrod, J. R. MacDonald, and H. E. Duckworth, Can. J. Phys. **43**, 275 (1965).
[21] B. Fastrup, P. Hvelplund, and C. A. Sautter, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. **35**, 10 (1966).
[22] P. Hvelplund and B. Fastrup, Phys. Rev. **165**, 405 (1968).
[23] G. W. Carriveau, G. Beauchemin, E. J. Knystautas, E. H. Pinnington, and R. Drouin, Phys. Lett. **46A**, 4 (1973).
[24] I. Hoffmann, E. Jäger, and U. Müller-Jahreis, Radiat. Eff. **31**, 57 (1976).
[25] P. Mertens, Nucl. Instrum. Methods **149**, 149 (1978).
[26] D. C. Santry and R. D. Werner, IEEE Trans. Nucl. Sci. **NS-26**, 1335 (1979).
[27] D. Ward, H. R. Andrews, I. R. Mitchell, W. N. Lennard, R. B. Walker, and N. Rud, Can. J. Phys. **57**, 645 (1979).
[28] F. Schulz and W. Brandt, Phys. Rev. B **26**, 4864 (1982).
[29] D. J. Land, D. G. Simons, J. G. Brennan, and G. A. Glass, Nucl. Instrum. Methods Phys. Res. B **10/11**, 234 (1985).
[30] D. C. Santry and R. D. Werner, Nucl. Instrum. Methods Phys. Res. B **53**, 7 (1991).
[31] D. S. Gemmell, Nucl. Instrum. Methods Phys. Res. **194**, 255 (1982).
[32] R. Garcia-Molina, C. D. Denton, I. Abril, and N. R. Arista, Phys. Rev. A **62**, 012901 (2000).
[33] V. V. Balashov, *Interaction of Particles and Radiation with Matter* (Springer, Berlin, 1997).