I. INTRODUCTION

The study of the differences between the energy loss of a single atomic ion or an ensemble of atomic ions is an interesting topic, which was first analyzed both theoretically and experimentally by Brandt et al. [1]; these differences, denominated vicinage effects in the energy loss, 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 vicinage effects.

Several works have been devoted to analyze experimentally the energy loss of different molecular projectiles, ranging from the simplest case H$_2^+$ [1], to more complex ones, such as O$_2^-$ [2], N$_2^+$, and O$_2^+$ [3], C$_n^+$ ($n=60$) [4–7], O$_2$, B$_3$, and C$_n$ ($n=2,3$) neutral molecules [8], B$_n^+$ ($n=2–4$) [9], and Si$_n^+$ ($n=3$) [10]. Most of such studies have shown that the energy lost by the atomic ions resulting from the dissociation of the molecular projectile, randomly oriented with respect to the beam direction, was higher than that of the same but isolated carbon atomic ions, which is small for velocities $v \sim 1$ a.u. and becomes more important for higher velocities ($v \sim 4$ a.u.). The energy loss of the dissociated atomic ions generally increases with the size and packing level of the cluster, although in some cases it tends to a saturation value (when the number of constituents of the cluster increases) or it could even decrease with cluster size for certain situations (for projectiles with $v \sim 1$ a.u. in aluminum or silicon targets). The vicinage effects in the energy loss also depend on the target nature, being more important for silicon and aluminum foils than for amorphous carbon foils. Our results show that in most cases the highest enhancement in energy loss should be expected for large clusters with high projectile velocities in aluminum or silicon targets. The experimental energy loss measured in carbon targets is well reproduced by our calculations.
our conclusions in Sec. IV. Atomic units will be used throughout this work except where otherwise stated.

II. SIMULATION CODE

Our simulation code [29,30] is based in a combination of molecular dynamics and Monte Carlo techniques. The molecular dynamics treatment consists in numerically solving Newton’s equation of motion for all particles that form the system we are analyzing. If we know at the time \( t \) the position, the velocity, and the forces that act on each particle moving through a solid, we can calculate their positions and velocities after a time step \( \Delta t \) and, therefore, at any time \( t + \Delta t \), where \( n \) is the number of time steps. Molecular dynamics is a deterministic technique but the capture and loss of electrons, the stopping force, and the nuclear scattering, which the projectile suffers, have a stochastic nature. Moreover, each atomic ion moving through a solid the atomic ions lose –loss processes until they reach a dynamical equilibrium.

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A. Stopping force

When the \( i \)-atomic ion (with atomic number \( Z_i \)) is moving with velocity \( v_j \) through a solid characterized by an energy-loss function \( \text{Im}[-1/e(k, \omega)] \), it is slowed down by losing energy in electronic excitations of the target (with energy \( \omega \) and momentum \( k \)); \( e(k, \omega) \) is the target dielectric function. This slowing down is characterized by the stopping power

\[
S_{p,i} = \frac{2}{\pi v_j^2} \int_0^{\infty} dk \left[ \frac{Z_i - \rho_i(k)}{k} \right] \text{Im} \left[ \frac{-1}{e(k, \omega)} \right] \Omega_i \omega^2 d\omega
\]

and the energy-loss straggling

\[
\Omega_i^2 = \frac{2}{\pi v_j^2} \int_0^{\infty} dk \left[ \frac{Z_i - \rho_i(k)}{k} \right] \text{Im} \left[ \frac{-1}{e(k, \omega)} \right] \Omega_i \omega^2 d\omega
\]

where \( \rho_i(k) \) is the Fourier transform of the electronic density of the \( i \)-atomic ion. The stopping force that acts on the \( i \)-atomic ion fluctuates because of the stochastic nature of the interaction with the target electrons. So the modulus of the stopping force, \( F_{\text{stopping,}i} \), is given by a Gaussian distribution, for which \( S_{p,i} \) is the mean value and \( \Omega_i/\Delta \) is the standard deviation; \( \Delta = v_0 \Delta t \) is the distance traveled by this atomic ion in a time step \( \Delta t \). Then

\[
F_{\text{stopping,}i} = -(S_{p,i} + (\Omega_i/\Delta) \xi_i) \hat{v}_i,
\]

where \( \xi_i \) is a random number obtained according to a Gaussian distribution [32] and \( \hat{v}_i \) is the unit vector of the instantaneous velocity \( v_i \).

For medium-size ions, like carbon, \( \rho_i(k) \) can be obtained for \( N_i \) bound electrons using the Brandt-Kitagawa model [33],

\[
\rho_i(k) = \frac{N_i}{1 + (k \Lambda_i)^2},
\]

where \( \Lambda_i = 0.48N_i^{2/3}/(Z_i - N_i/7) \) being a variational parameter that minimizes the internal energy of the atomic ion; this result practically coincides with that obtained from the Lenz-Jensen model [33]. The Brandt-Kitagawa model [33] is one of the most used descriptions for the electronic charge density of the projectile, because of the straightforwardness of the analytical expressions obtained for \( \rho_i(k) \) and the good comparison with experimental data [34].

A realistic description of \( \text{Im}[-1/e(k, \omega)] \) is obtained by combining a sum of Mermin-type [35–39] energy-loss functions that fits to experimental optical data and the use of generalized oscillator strengths [37–39]; these contributions account for excitations of the target outer-shell and inner-shell electrons, respectively.

B. Wake force and Coulomb repulsion

The electronic excitations created in the target by a projectile also affect the motion of its partners, through the so-called wake force [1,40,41]. This force is calculated as stated in Ref. [36] for protons, but now using the Brandt-Kitagawa model [33] to describe the electrons bound to each atomic ion. Using cylindrical coordinates, the wake force acting on the \( i \)-atomic ion due to its neighbor, denoted by \( j \), can be decomposed into its parallel and perpendicular components with respect to the velocity \( v_j \),
The components of the internuclear vector \( \mathbf{R}_{ij} \) are parallel and perpendicular to the velocity \( \mathbf{v}_i \), acting on the \( i \)-atomic ion due to its neighbor, denoted by \( j \). \( \mathbf{R}_{ij} \) and \( \mathbf{R}_{ij} \) are the parallel and perpendicular components of the internuclear vector \( \mathbf{R}_{ij} \) between the \( i \)- and \( j \)-atomic ions.

\[
F_{ij} = \frac{2}{\pi v_j} \int_0^\infty \frac{dk}{k} \left( Z_i - \rho_i(k) \right) \left( Z_j - \rho_j(k) \right) \times \int_0^{k_{ij}} d\omega \omega \sqrt{k^2 - \omega^2 v_j^2} \cos \left( \frac{\omega R_{ij}}{v_j} \right) \left[ \frac{1}{\epsilon(k, \omega)} - 1 \right] + \sin \left( \frac{\omega R_{ij}}{v_j} \right) \left[ \frac{1}{\epsilon(k, \omega)} - 1 \right]
\]

and

\[
F_{ij} = \frac{2}{\pi v_j} \int_0^\infty \frac{dk}{k} \left( Z_i - \rho_i(k) \right) \left( Z_j - \rho_j(k) \right) \times \int_0^{k_{ij}} d\omega \omega \sqrt{k^2 - \omega^2 v_j^2} \sin \left( \frac{\omega R_{ij}}{v_j} \right) \left[ \frac{1}{\epsilon(k, \omega)} - 1 \right] + \cos \left( \frac{\omega R_{ij}}{v_j} \right) \left[ \frac{1}{\epsilon(k, \omega)} - 1 \right]
\]

where \( R_{ij} \) and \( R_{ij} \) are the parallel and perpendicular components of the internuclear vector \( \mathbf{R}_{ij} \) between the \( i \)- and \( j \)-atomic ions (see Fig. 1); \( J_0(\cdot) \) and \( J_1(\cdot) \) are Bessel functions of the first kind [42].

To simplify the simulations we evaluate Eqs. (6) and (7) using the same velocity for all atomic ions that form the molecular projectile. This is a plausible approximation because the velocities of the atomic ions only become significantly different for large depths inside the target, but in such situations the wake forces are negligible due to the large distance between the molecular constituents, because of the Coulomb repulsion. Leaving aside the evaluation of Eqs. (6) and (7), the different velocities of the atomic ions are properly considered through all the simulation.

The total wake force that acts on the \( i \)-atomic ion due to all its neighbors is given by

\[
F_{\text{wake},i} = \sum_{j \neq i} \sqrt{F_{ij}^2 + F_{ij}^2} \mathbf{v}_j,
\]

where \( n \) is the number of atomic ions that constitutes the molecular projectile. It is important to note that for a given target, the stopping force depends on the projectile velocity and charge state, Eqs. (2)–(4), but the wake force depends also on the positions and velocities of the rest of the atomic ions resulting from the cluster dissociation, Eqs. (6)–(8).

The mutual repulsion between charged neighbors is considered by means of a pure Coulomb potential, which gives the following force acting on the \( i \)-atomic ion:

\[
F_{\text{Coulomb},i} = \left( Z_i - N_i \right) \sum_{j \neq i} \frac{q_i q_j}{R_{ij}^2} \mathbf{R}_{ij} = q_i q_j \sum_{j \neq i} R_{ij} \mathbf{R}_{ij},
\]

where \( q_i = Z_i - N_i \) is the charge state of the \( i \)-atomic ion (analogously for the \( j \)-atomic ion) and \( \mathbf{R}_{ij} \) is the unit vector of \( \mathbf{R}_{ij} \).

### C. Nuclear scattering

Multiple scattering of an atomic ion with the target atomic cores is considered through a Monte Carlo model [43,44], using the universal potential cross section [34] to calculate the scattering angles and the corresponding nuclear energy loss. In this treatment the reduced path length between two successive collisions is given by

\[
L = -\frac{4\mu}{m_1 + m_2} \ln(\xi_1),
\]

where \( \xi_1 \) is a random number uniformly distributed between 0 and 1; \( m_1 \) and \( m_2 \) are the projectile and target atomic masses, respectively; \( \mu \) is the reduced mass, and \( J_{\text{tot}} = 1/(4N_{\text{tot}}a_{\text{U}}^2) \) is an estimation of the reduced total cross section, \( N_{\text{tot}} \) being the target atomic density and \( a_{\text{U}} \) the universal screening length [34],

\[
a_{\text{U}} = \frac{0.8853}{Z_i^{0.23} + Z_j^{0.23}},
\]

where \( Z_i \) and \( Z_j \) are the projectile and target atomic numbers. The path length \( L \) between two successive collisions is directly related to \( L \) by means of

\[
L = \frac{m_1 + m_2}{4\pi a_{\text{U}}^2 \mu k N L}.
\]

The polar scattering angle \( \theta \) relative to the ion direction of motion is given as a function of the parameter \( \eta \) through

\[
\cos \theta = \left( 1 - \frac{2\mu \eta}{m_1 c^2} \left[ 1 - \frac{4\mu \eta}{(m_1 + m_2)c^2} \right]^{-1/2} \right),
\]

where \( \mathcal{E} \) is the reduced energy defined by

\[
\mathcal{E} = \frac{a_{\text{U}} m_2}{Z_i Z_j (m_1 + m_2)} E,
\]

\( E \) being the projectile instantaneous energy.
THE VALUE OF THE PARAMETER $\eta$ IS CALCULATED USING

$$J(\eta) = J(\xi) + \xi_2 J_{\text{tot}},$$

(15)

where $\xi_2$ is a random number, uniformly distributed between 0 and 1. The scattering cross section in reduced units, $J(\eta)$, can be evaluated using

$$J(\eta) = J(\eta_0) + \int_{\eta_0}^{\eta} d\eta' \frac{f(\eta')}{\eta'^2},$$

(16)

where $f(\eta)$ is the function given by Meyer [45] and we assume $\eta_0 = 10^{-3}$ as a fixed lower integration limit. Equation (15) is independent of $\eta_0$, because both $J(\eta)$ and $J(\xi)$ depend on $\eta_0$ through Eq. (16).

Finally, the azimuthal scattering angle $\psi$ relative to the ion direction of motion is evaluated as

$$\psi = 2\pi \xi_3,$$

(17)

where $\xi_3$ is a random number uniformly distributed between 0 and 1.

Figure 2 sketches the motion of an atomic ion before the $m$ collision, in a direction defined by the polar angle $\Theta_{m-1}$ and the azimuthal angle $\Psi_{m-1}$ in the laboratory frame of reference. We obtain $L$ by means of Eq. (12); after an elapsed time $L/v$, the $m$ collision takes place and we determine the scattering angles $\theta$ and $\psi$ using the Monte Carlo technique. The new direction of the projectile after the $m$ collision is defined by [44,46]

$$\cos \Theta_m = -\sin \theta \cos \psi \sin \Theta_{m-1} + \cos \theta \cos \Theta_{m-1},$$

$$\cos \Psi_m = \frac{1}{\sin \Theta_m} (\sin \theta \cos \psi \cos \Theta_{m-1} \cos \Psi_{m-1}$$

$$- \sin \theta \sin \psi \sin \Psi_{m-1} + \cos \theta \sin \Theta_{m-1} \cos \Psi_{m-1}).$$

(18)

D. Electron capture and loss

The capture and loss of electrons by the dissociated atomic ions have been incorporated in our simulation code using a model similar to that suggested in Ref. [44]. We assume that the electron-loss cross section $\sigma_{\text{loss},i}$ for the $i$-atomic ion is proportional to the number of bound electrons $N_i = Z_i - q_i$,

$$\sigma_{\text{loss},i}(q_i \rightarrow q_i + 1) = (Z_i - q_i)\sigma_i,$$

(19)

where $q_i \rightarrow q_i + 1$ denotes that the atomic ion changes its charge state from $q_i$ to $q_i + 1$ and $\sigma_i$ is the geometrical cross section of the $i$-atomic ion,

$$\sigma_i = \pi \langle r^2 \rangle = 4\pi \Lambda^2,$$

(20)

with $\langle r^2 \rangle = 2\Lambda_i$ being the average distance between the bound electrons and the atomic nucleus in the Brandt-Kitagawa model [33].

The inverse mean free path for electron loss is

$$\mu_{\text{loss},i}(q_i \rightarrow q_i + 1) = N \sigma_{\text{loss},i}(q_i \rightarrow q_i + 1).$$

(21)

If multiple-electron processes are neglected, the inverse mean free path for electron capture from the equilibrium relation is

$$\mu_{\text{capt},i}(q_i + 1 \rightarrow q_i) = \frac{\phi(q_i)}{\phi(q_i + 1)} \mu_{\text{loss},i}(q_i \rightarrow q_i + 1),$$

(22)

where $\phi(q)$ and $\phi(q+1)$ are the equilibrium fractions of the $q_i$ and $q_i + 1$ charge states, respectively. Then the probability of electron loss, $P_{\text{loss},i}$ (or electron capture, $P_{\text{capt},i}$), by the $i$-atomic ion with charge state $q_i$ is

$$P_{\text{loss},i}(q_i \rightarrow q_i + 1) = \frac{\mu_{\text{loss},i}(q_i \rightarrow q_i + 1)}{\mu_{\text{capt},i}(q_i + 1 \rightarrow q_i) + \mu_{\text{loss},i}(q_i \rightarrow q_i + 1)},$$

(23)

$$P_{\text{capt},i}(q_i + 1 \rightarrow q_i) = 1 - P_{\text{loss},i}(q_i \rightarrow q_i + 1).$$

(24)

According to these probabilities our simulation code chooses at each time step whether the $i$-atomic ion loses or captures an electron.

To satisfy the normalization condition $\sum_{q_i=0}^{\infty} \phi(q_i) = 1$, each charge state fraction $\phi(q_i)$ of the $i$-atomic ion is evaluated through

$$\phi(q_i) = \frac{f(q_i)}{\sum_{q_i=0}^{\infty} f(q_i)},$$

(25)

where the distribution of charge states with $\langle q_i \rangle$ mean equilibrium charge state and $\sigma_i$ standard deviation is given by

$$f(q_i) = \frac{1}{\sqrt{2\pi \sigma_i^2}} \exp \left[ -\frac{(q_i - \langle q_i \rangle)^2}{2\sigma_i^2} \right].$$

(26)

Both $\langle q_i \rangle$ and $\sigma_i$ are obtained through a fit to experimental data [47] and depend on $Z_i$, $v_i$, and on the target atomic number $Z$.

III. RESULTS

Taking into account all the ingredients presented above, we can obtain the coordinates and velocities of any atomic
observed in Fig. 3 is reached. C5+, with the probability of finding the atomic ions in an

distribution, which is independent of the transient time needed to acquire the equilibrium charge-state distributions coincide with the ones given by the CASP code [49]; the thin line curves labeled 0, +1, +2, . . . , +6 are the contributions to the stopping power due to each charge state of the carbon ion. The inset shows the calculated energy-loss straggling per unit path length, using Eqs. (3) and (28).

The charge state of a swift atomic ion moving through a solid is characterized by the stopping power and the energy loss straggling [48]

\[
S_p = \sum_{q=0}^{Z_i} \phi(q) S_{p,i}, \tag{27}
\]

\[
\Omega^2 = \sum_{q=0}^{Z_i} \phi(q) \Omega^2_i, \tag{28}
\]

where \(S_{p,i}\) and \(\Omega^2_i\) come from Eqs. (2) and (3), respectively, and the charge fractions are derived from Eq. (25). Figure 4 depicts \(S_p\) and \(\Omega^2\) of amorphous carbon for swift carbon ions. The symbols are experimental data [49], and the thick solid lines correspond to our calculations. The contributions to the stopping power due to the different charge states \(q_i\) of the projectile—i.e., \(\phi(q_i)S_{p,i}\)—are represented by different types of thin lines, the labels indicating the corresponding charge state. Similar results were obtained for silicon and aluminum targets. In all cases considered in this work there is a satisfactory agreement between our calculations and the experimental data in a broad range of projectile velocities.

B. Dissociated atomic ions

The charge state of a swift atomic ion moving through a solid is different when it is isolated or when it forms part of a molecular projectile. In general, for a given velocity the average charge state of a dissociated atomic ion is lower than...
that of an isolated atomic ion and it depends on the molecular structure and velocity, as well as on the foil composition and thickness \[17,50–57\]. These vicinage effects in the charge state are related to the proximity of the neighboring atomic ions as they move through the target.

Recent experiments \[53,55\] show that dissociated atomic ions have smaller charge state than the corresponding isolated ions. We obtained in Ref. \[57\] that the reduction in charge state inside the target is lower than that experimentally measured just at the exit of the foil. That reduction scarcely affects the energy loss of the dissociated atomic ions \[58\]. Therefore we assume in this work that the charge state of each atomic ion resulting from the molecular dissociation is identical to that corresponding to the same atomic (but isolated) ion.

The energy loss of the atomic ions resulting from the dissociation of a cluster depends on its size, geometry, and velocity, as well as on the target nature. The differences between the energy loss of a cluster and the sum of the energy loss of each one of its constituents moving with the same velocity but isolated—namely, the vicinage effects in the energy loss—are quantified through the stopping power ratio

\[
R = \frac{\Delta E(C_n^+)}{n\Delta E(C^+)},
\]

where \(\Delta E(C_n^+)\) and \(\Delta E(C^+)\) are the energy lost by the \(C_n^+\) cluster and an isostatic \(C^+\) ion, respectively; \(n\) is the number of atoms that form the cluster.

To analyze the dependence of the vicinage effects in the energy loss with the projectile size and geometrical structure, we have calculated the energy loss of \(C_n^+\) clusters \((n = 2–60)\) moving through amorphous carbon, aluminum, and silicon foils. These values of \(n\) cover wide types of geometries: linear \((n = 2–8)\) \[59–63\], ring-shaped \((n = 3, 10)\) \[64\], and cagelike structures \((n = 20)\) \[64,65\]. The geometrical structures of a few representative clusters are depicted in Fig. 5.

We show in Fig. 6 the stopping power ratio \(R\) as a function of the target thickness \(d\) for \(C_n^+\) \((n = 3, 10, 20, 36, \text{ and } 60)\) clusters incident with velocities \(v = 1, 2, 3, \text{ and } 4\) a.u. on amorphous carbon, aluminum, and silicon foils. A linear structure for the \(C_3^+\) has been assumed in order to analyze the transition between the different geometries. Curves corresponding to \(C_n^+\) ions with \(n = 2, 4–8, 28, \text{ and } 50\) have been omitted for clarity reasons, since they do not report any additional feature.

For almost all cluster sizes, velocities, and foil thicknesses, \(R > 1\). This trend is not satisfied for the thinner foils, where \(R < 1\). Such behavior, more noticeable for low \(v\), could be attributed to the different initial charge states of the \(C^+\) as compared to those of the dissociated atomic ions \((n = 1)\) neutral C atoms and one \(C^+\) ion, for a \(C_n^+\) cluster). In addition, \(R \rightarrow 1\) the thicker is the foil and the lower is the velocity, so the vicinage effects in the energy loss decrease when the dwell time increases. This is due to two factors that increase the interatomic distances and, therefore, distort the correlated motion of the cluster fragments: (i) the nuclear scattering (more important for the thicker foils) and (ii) the Coulomb explosion (more significant for the thinner foils). The vicinage effects in the energy loss also depend on the cluster velocity \(v\); the value of \(R\) clearly increases with \(v\). For instance, \(R \approx 3\) for \(C_{60}^+\) moving with \(v = 4\) a.u. through \(\approx 500\)-a.u. aluminum foil. Moreover, the energy-loss enhancement of the \(C_n^+\) clusters strongly depends on the target nature, being more important for silicon or aluminum foils than for amorphous carbon. This behavior is due to the larger extension of the wake forces induced in silicon and aluminum compared with the ones in amorphous carbon, as can be seen in Fig. 7 and in Refs. \[36,66\].

On the other hand, for amorphous carbon foils there is a sizable increase in the stopping power ratio with the cluster size for \(n = 3–20\), which is due not only to the increase of \(n\), but also to the change of structure (linear, ring-shaped, or cagelike); the combined effect of both factors increases the number of neighbors around each atomic ion, with the consequent enhancement of interactions, and the corresponding vicinage effects, felt by the atomic ions. Figure 6 shows no important differences in value of \(R\) for the case of \(C_n^+\) clusters in amorphous carbon foils when \(n\) increases even more \((n = 20–60)\). We have also observed this tendency for \(C_{28}^+\) and \(C_{50}^+\) ions. The absence of a significant variation for the stopping power ratio \(R\) in this case can be explained in terms of cancellations of vicinage effects due to the presence of several atomic ions at different distances from a given atomic ion.

A deviation of this general trend is observed for silicon and aluminum foils; for instance, \(R(C_3^+) > R(C_{n}^+)\) with \(v = 1\) a.u., whatever the value of \(n\) was. This anomaly is also observed for \(C_{20}^+\) with \(v = 2\) a.u. In addition, it is worth noticing for such targets the different shape of the \(R\) curves for
ENERGY-LOSS CALCULATION OF SWIFT $C_n^+$ (n...)

The cluster size and their velocities and sizes, as a function of the foil thickness $d$. The cluster velocity and target material are indicated as different rows and columns, respectively. The different types of curves indicate the cluster size: $n=3$ (solid lines), $n=10$ (dashed lines), $n=20$ (dotted lines), $n=36$ (dot-dashed lines), and $n=60$ (gray lines).

$C_{60}^+$ ions in relation to those for $C_n^+$ ($n=3, 10, 20$, and $36$) ions, both with velocity $v=2$ a.u.; the slope of the stopping power ratio clearly changes for $C_{60}^+$ when $200 \leq d \leq 800$ a.u. This change of slope also is observed for $C_{50}^+$ ions.

To explain this behavior we show in Fig. 7 the wake forces $F_\parallel$ and $F_\perp$ that act on a $C^{2+}$ ion (leading ion) due to another $C^{2+}$ ion (partner ion) as a function of the distance $R_0$, both moving with $v=2$ a.u. through amorphous carbon or silicon foils. We have chosen the most probable charge state ($q=2$) for $v=2$ a.u. [see Fig. 3(b)]. We depict the distribution of interatomic distances for $C_{60}^+$ at $d=500$ a.u. (where the change in the slope of $R$ is more pronounced) to help us in determining the most probable interatomic distances between atomic ions. The wake forces $F_\parallel$ and $F_\perp$ are shown for $0.25 \leq R_0 \leq 5$ a.u. because the more important contribution to the total wake force felt by the partner ion comes from such small values of $R_0$ (contributions coming from larger values of $R_0$ can be neglected in a qualitative discussion).

According to the figure, $F_\parallel < 0$ and show a similar behavior in front of the leading ion for both amorphous carbon and silicon targets. On the contrary, differences on the wake forces appear behind the leading ion: generally $F_\parallel > 0$ for amorphous carbon, whereas $F_\parallel$ for silicon can have both positive and negative values. In addition, $F_\perp < 0$ for amorphous carbon, whatever the partner ion is, but for silicon targets it can be positive or negative. Such negative values of $F_\perp$ for amorphous carbon tend to align the partner ions, whereas the positive values of $F_\perp$ for silicon give rise to an nonalignment effect. So the wake forces $F_\parallel$ and $F_\perp$ that act on the partner ion can be positive or negative behind the leading ion for silicon targets, compensating themselves and giving rise to cancellation effects in the energy loss; such a cancellation is not observed for amorphous carbon foils, because almost always $F_\parallel > 0$ or $F_\perp < 0$ behind the leading ion, as can be noticed in Fig. 7. These cancellation effects are responsible for the observed reduction of $R$ for $C_{60}^+$ in silicon when $200 \leq d \leq 800$ a.u.

The dependence of $R$ on the cluster size $n$ is illustrated in Fig. 8 for $C_n^+$ ($n=2-60$) clusters moving through several foil thicknesses. The same graphs are depicted in the insets, but without breaking the horizontal axis; the scale of the vertical axis is identical to those of the corresponding main graph. The purpose of these insets is to show more clearly the dependence on $n$. For amorphous carbon foils $R$ increases with $n$ for the smaller clusters ($n=2-20$), but it remains almost constant for the larger clusters ($n=20-60$). However, for silicon and aluminum foils there is a maximum of $R$ for $C_3^+$ with $v=1$ a.u. and for $C_{20}^+$ with $v=2$ a.u., which only is observed for the thinner foils ($d=250$ a.u.); the maximum of the stopping power ratio is moved to larger clusters as the velocity increases. This anomaly, which can

FIG. 6. Stopping power ratio $R$ for several target materials, cluster velocities, and sizes, as a function of the foil thickness $d$. The cluster velocity and target material are indicated as different rows and columns, respectively. The different types of curves indicate the cluster size: $n=3$ (solid lines), $n=10$ (dashed lines), $n=20$ (dotted lines), $n=36$ (dot-dashed lines), and $n=60$ (gray lines).

FIG. 7. Left axis: parallel and perpendicular components of the wake forces that act on a $C^{2+}$ ion due to a neighbor $C^{2+}$ ion (located at the origin of the graph, $R_0=0$), as a function of the parallel component $R_0$ of the interatomic distance; both ions moving with $v=2$ a.u. through amorphous carbon or silicon foils. The meaning of the lines are (solid line) $F_\parallel$ for $R_0=0$, (dashed line) $F_\parallel$ for $R_0=0.25$ a.u., (dotted line) $F_\parallel$ for $R_0=5$ a.u., and (dot-dashed line) $F_\parallel$ for $R_0=5$ a.u. Right axis: the interatomic distance distributions of $C_{60}^+$ at $d=500$ a.u. are depicted as shaded histograms.
be explained according to the reasoning presented above, is not observed for thicker foils, where the stopping power ratio increases with the number \( n \) of constituents.

In conclusion, the number of atomic ions and the geometrical structure of the cluster affect the vicinage effects in the energy loss. In general one could expect that \( R \) increases with the size \( n \) of the cluster. However, this tendency cannot be observed for the larger clusters due to the cancellation of the vicinage effects, which arises from the different inter-atomic distances of the atomic ions that produce positive and/or negative vicinage effects in the energy loss [67].

C. Comparison with experimental data

Baudin et al. [4,6] measured the energy loss of \( C_n^+ (n = 2−8) \) ions incident with energy \( E_0 = 1.01−5.65 \) MeV/atom on 250-Å-thick amorphous carbon foils, finding slight vicinage effects in the energy loss of the molecular projectiles compared to the same but isolated atomic ions. However, they did not detect (compatible with their experimental resolution of 5%) vicinage effects for \( C_{60}^+ \) clusters incident with \( E_0 = 305 \) and 455 keV/atom on 300- and 500-Å-thick amorphous carbon foils. Tomaschko et al. [5] also reported measurements on the energy loss of small carbon clusters \( C_n^+ (n = 2−5) \) incident with \( E_0 = 1.4−4.0 \) MeV on amorphous carbon, Formvar, and gold foils. Contrary to what was observed by Baudin et al. [4,6], they did not detect vicinage effects in the energy loss, according to the error bars of their measurements. Besides the stopping power ratio, in order to quantify vicinage effects in the energy loss we use the difference between the mean energy loss of each molecular fragment resulting from the dissociation of \( C_n^+ \) and that of the isolated atomic ions,

FIG. 8. Stopping power ratio \( R \) as a function of the number \( n \) of atomic ions that constitutes the cluster, for different projectile velocities and target materials: (□) amorphous carbon, (○) aluminum, and (△) silicon. The target thickness \( d \) and projectile velocity \( v \) are indicated in the figure. The lines joining the symbols are drawn only to guide the eye. The insets show the behavior of \( R \) without the break in the horizontal axis; note that the vertical scale in each inset coincides with that in the main graph.
FIG. 9. Difference $D$ between the mean energy loss of each molecular fragment resulting from the dissociation of a $C_n^+$ ($n = 2–8$) ion and that of the isolated carbon atomic ions, as a function of the number $n$ of atomic ions that constitutes the cluster, incident with different projectile energies $E_0$ (indicated in the legend of the figure) on 250-Å-thick amorphous carbon foils. The open circles (and the error bars) represent the experimental data [4,6], whereas gray triangles correspond to our results. The crosses represent the results for the $C_3^+$ cluster when we assume a linear structure.

$$D = \frac{\Delta E(C_n^+)}{n} - \Delta E(C^+).$$  \hspace{1cm} (30)

In Fig. 9 we show $D$ for small $C_n^+$ ($n = 2–8$) ions incident with energy $E_0 = 1.01 - 5.65$ MeV/atom on 250-Å-thick amorphous carbon foils. We have done our simulations using the geometrical structures of the $C_n^+$ ($n = 2–8$) ions in the ground state: bent for $C_3^+$ [64] and linear for the rest of the molecular ions [59–63]. The open circles and the corresponding error bars represent the experimental data [4,6], whereas gray triangles correspond to our results. The crosses represent the corresponding results when we assume a linear structure for the $C_3^+$ cluster.

An enhancement in the energy loss of the cluster compared to that of its isolated constituents is observed for $E_0 \approx 2$ MeV/atom. The agreement of our calculations and the experimental data is fairly good for all the experimental situations analyzed, except for the lowest projectile energy ($E_0 = 1.01$ MeV/atom), where the behavior shown by these measurements [6] differs significantly from the rest of experimental data [4].

When considering in our calculations a linear structure for the $C_3^+$ ion small differences in $D$ (lower than 8%) appear, both being calculations (with a triangular or a linear structure) within the error bars; so from these results it is not possible to state the geometry of the cluster in the experiment. The Coulomb explosion imaging technique [41,68] or the charge-state distributions of the dissociated atomic ions at the exit of the target [53,55–57,69] could be used for this purpose.

As expected, $D$ increases with the number $n$ of atomic constituents and also grows as the projectile energy becomes higher. Such a dependence with the projectile energy explains why no enhancement in the energy loss was observed by Tomaschko et al. [5], since the projectile energies in their experiments were lower than those of the projectiles used by Baudin et al. [4].

Recently, Kaneko [17] has reported an analytical calculation of the energy loss of swift $C_n^+$ ($n = 2–8$) ions incident on amorphous carbon foils, obtaining a good agreement with the experimental data [4,6]. According to that model, the electron binding energy of an atomic ion that is part of an ensemble of correlated atomic ions is higher than for an isolated atomic projectile [17]. This effect was evaluated using a pure Coulomb potential for the interaction between the dissociated atomic ions [17], which does not seem a plausible approximation because screening by the target electrons is not considered [56,57]. In addition, these calculations did not take into account the multiple scattering with the target nuclei or how the wake forces affect the motion of the dissociated atomic ions [17]; those effects have been proven to be determinant to explain the correlated motion of the dissociated fragments [41]. The variation in the charge state of the dissociated atomic ions before the equilibrium is acquired (i.e., the transient charge state) also was neglected [17], although the need for considering that effect was previously reported when evaluating the energy loss of swift molecular projectiles traversing thin foils [12,16]. On the contrary, our simulation code includes multiple scattering with the target nuclei, the wake forces, and the transient charge state; it does not include vicinage effects in the charge state, but as we have previously stated, these effects only represent a decrease of less than 5% in the stopping power ratio for the most unfavorable case discussed in this work.

Concerning the experiment performed for $C_{60}^+$ incident with $E_0 = 305$ and 455 keV/atom on 300- and 500-Å-thick amorphous carbon foils [4], we depict in Fig. 10 the energy distribution of the dissociated atomic ions compared with that of isolated carbon atomic ions: they are plotted just at the exit of the foil, not when the particles reach a far detector, because Coulomb explosion (the only interaction) during the travel to the detector broadens the energy distribution without changing its mean energy. Two main features can be seen in these distributions: the ones corresponding to dissociated atomic ions are almost two times wider, and with a smaller mean energy, than the ones for isolated carbon atomic ions. The broadening of the energy distribution can be understood in terms of Coulomb repulsion inside the foil, which accelerates the leading fragments and slows down the trailing ones.
energy per atomic ion, for the two foil thicknesses. On the other hand, as expected, decreases with the foil thickness and tends to one, the thicker the foil. On the other hand, grows for the thicker foils, but it is very small, being 1% of the initial energy per atomic ion, for the two foil thicknesses (300 and 500 Å) corresponding to the experimental data [4], and it slowly grows as the foil gets thicker. Therefore, our simulations predict a small enhancement in the energy loss of the dissociated atomic ions in comparison with the isolated ones, which is in good agreement with the experimental observations [4]. All the same, the behavior of both curves and show that vicinage effects in the energy loss decrease when the foil thickness increases.

IV. CONCLUSIONS

We have calculated the energy loss of swift $C_n^+$ ($n = 2–60$) clusters moving with velocities $1 \leq v \leq 4$ a.u. in amorphous carbon, aluminum, and silicon foils. Our simulations predict $R > 1$ in most of the cases analyzed. These vicinage effects in the energy loss usually increase with the projectile size and velocity and also depend on the target nature, being more important for silicon and aluminum foils than for amorphous carbon foils. Then, molecular projectiles incident with high energy in silicon or aluminum thin foils should be used in order to measure the highest enhancement in their energy loss with respect to the corresponding atomic ions. Projectiles with high energy loss could be of interest in inertial confinement fusion.

Figure 11 shows $R$ and $D$ as a function of the foil thickness for two incident energies: $E_0 = 305$ and 455 keV/atom. As expected, $R$ decreases with the foil thickness and tends to one, the thicker the foil. On the other hand, $D$ grows for the thicker foils, but it is very small, being $\leq 1\%$ of the initial energy for the two foil thicknesses (300 and 455 keV/atom). Nevertheless, our simulations only predict a small enhancement ($\approx 1\%$) in the energy loss of $C_n^+$ clusters incident with $v \approx 1$ a.u. in amorphous carbon foils of 300 and 500 Å thicknesses, in accordance with the experimental observation, where no energy-loss enhancement was detected within 5% of experimental limit of observation [4]. This lack of sizable energy-loss differences between fragments of large molecules or clusters and individual atomic ions may be understood in terms of the loss of spatial correlation of the former due to Coulomb repulsion and nuclear scattering, which increase the interatomic distances between the dissociated fragments.

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