Monte Carlo calculations of the energy loss for H_2^+ molecular ions transmitted through thin C and Al foils

Mario M. Jakas and Néstor E. Capuj
Departamento de Física Fundamental y Experimental, Universidad de La Laguna, 38201 La Laguna, Tenerife, Spain
(Received 31 October 1994)

We calculate the mean energy loss of H_2^+ molecular ions transmitted through carbon and aluminum foils as a function of the bombarding energy. Our results are in good agreement with available experimental data. Furthermore, inhomogeneities containing the foil are found to be essential in the energy-loss measurement of transmitted molecular ions.

PACS number(s): 34.50.Bw, 79.20.Nc

I. INTRODUCTION

It is well known that the mean energy loss of fast molecular ions transmitted through thin solid films differs from that of the constituent nuclei at the same velocity. This phenomenon, known as the *vicinage effect* on the stopping, is not only well documented [1–9], but also the elementary processes that produce such an effect appear to be relatively well understood. According to the linear theory of stopping [10,11], the leading ion in the cluster induces an electron density fluctuation around the trailing ion. Such a fluctuation, which resembles the wake in the water produced by a moving boat, causes the stopping of the trailing ion to differ from that of an isolated proton at the same velocity. The stopping of the cluster therefore will differ from that of two *uncorrelated* protons by an amount that was found to depend on both the velocity and the relative position of the nuclei. It is thus clear that in order to calculate the mean energy loss of the transmitted molecules, one has to know the trajectory of the nuclei that produced these molecules. Unfortunately, however, as the passage of two correlated protons through thin films constitutes a very complicated problem, no such calculation has been reported so far. Furthermore, as transmission yields are usually too small, this problem cannot be easily calculated using standard Monte Carlo (MC) simulations.

Recently, the transmission yields of H_2^+ ions were calculated within computer times that were a small fraction of the time one would need using a standard MC calculation [12]. This enhanced efficiency was attained by means of special MC techniques that enabled our MC simulations to deal with rare events in a very efficient manner. Using such a numerical code, we report in this paper MC calculations of the mean energy loss for H_2^+ transmitted through carbon and aluminum foils. Our results are found to compare fairly well with previous experiments. In addition, these results can be explained in terms of the wake forces and the way transmission "selects" from among the different trajectories of the protons during passage through the foil.

The plan of this paper is as follows. In Sec. II we show the assumptions employed to calculate the mean energy loss for the two protons in the cluster. A detailed description of the MC code is, however, purposely omitted since it has been published elsewhere [12]. In Sec. III A the results of our MC calculations together with a comparison with previous experiments are given. The role of foil inhomogeneities in this type of measurement is investigated in Sec. III B. This relates to an early controversy about the extent to which inhomogeneities in the foil may distort stopping measurements of transmitted molecules [3,13]. A summary is contained in Sec. IV.

II. CALCULATION

During penetration, the center of mass (c.m.) of the H fragments are subjected to a force

$$\mathbf{F}_{\text{c.m.}}=\mathbf{F}_1+\mathbf{F}_2 , \tag{1}$$

where $\mathbf{F}_i$ is the force acting upon the ith nucleus, with $i=1,2$. These two forces can be separated into the components

$$\mathbf{F}_i=\mathbf{F}_i^{(w)}+\mathbf{F}_i^{(C)}+\mathbf{F}_i^{(\text{stop})} \quad \text{for } i=1,2 . \tag{2}$$

Here $\mathbf{F}^{(w)}$ stands for the wake force, $\mathbf{F}^{(C)}$ is the Coulomb repulsion acting upon the fragments, and $\mathbf{F}^{(\text{stop})}$ accounts for the *uncorrelated* stopping. The latter force is assumed to include a series of randomly *impulsive* forces simulating the energy-loss straggling.

The energy loss after dwell time τ in the foil becomes

$$\Delta E_{\text{c.m.}}(\tau)=-\int_0^\tau dt\ \mathbf{v}\cdot\mathbf{F}_{\text{c.m.}} . \tag{3}$$

Taking an average over different trajectories, one has

$$\langle \Delta E_{\text{c.m.}} \rangle(\tau)=-\int_0^\tau dt \langle \mathbf{v}\cdot\mathbf{F}_{\text{c.m.}} \rangle . \tag{4}$$

Since the Coulomb forces are *internal,* i.e., $\mathbf{F}_1^{(C)}=-\mathbf{F}_2^{(C)}$, they do not participate in modifying the molecule energy. Furthermore, the mean uncorrelated stopping of the two nuclei are, by definition, the same; hence we have

$$\langle \Delta E_{\text{c.m.}} \rangle(\tau)=2\langle \Delta E_p \rangle(\tau)+\langle \Delta E^{(w)} \rangle(\tau) , \tag{5}$$

where $\langle \Delta E_p \rangle$ is the mean energy loss of protons at the same velocity. Similarly, given that the wake force acts only upon the trailing ion in the cluster, we can finally

write

$$\Delta E^{(w)}(\tau) = -\int_0^{\tau} dt\, \mathbf{v}\cdot\mathbf{F}^{(\mathrm{trail})} . \tag{6}$$

Here $\mathbf{F}^{(\mathrm{trail})}$ represents the wake force acting upon the trailing proton in the cluster.

According to Eq. (5) the mean energy loss of transmitted molecules *relative* to that of two uncorrelated protons at the same velocity results

$$\frac{\langle \Delta E_{\mathrm{c.m.}} \rangle_{\mathrm{mol}}(\tau)}{2\langle \Delta E_p \rangle(\tau)} = 1 + \frac{\langle \Delta E^{(w)} \rangle(\tau)}{2\langle \Delta E_p \rangle(\tau)} , \tag{7}$$

where $\langle\ \rangle_{\mathrm{mol}}$ indicates an average over trajectories that produced molecules. Using the notation introduced by Laubert [3], we can rewrite the stopping ratio in Eq. (7) as

$$R_2 = \frac{\Delta E(H_2^+, H_2^+)}{2\Delta E(H^+, H^+)} = 1 + \frac{\langle \Delta E^{(w)} \rangle(\tau)}{2\langle \Delta E_p \rangle(\tau)} , \tag{8}$$

where ΔE(in, out) denotes the mean energy loss for particles arriving as "in" and detected as "out" after traversing the foil.

Using Monte Carlo simulations we calculate $\langle \Delta E^{(w)} \rangle(\tau)$ as a function of the bombarding energy, the dwell time in the foil, and two different targets C and Al. As the velocity of the protons during passage results change only slightly one can replace $\mathbf{v}$ by the initial velocity v_0. Hence, assuming that the z axis is along the beam direction we have

$$\Delta E^{(w)}(\tau) = v_0 |e| \int_0^{\tau} dt \frac{\partial \phi^{(w)}}{\partial z} , \tag{9}$$

where $\phi^{(w)}$ is the "oscillatory" part of the wake potential and e is the elementary charge. We thus calculated $\Delta E^{(w)}(\tau)$ according to Eq. (9) for every trajectory and then took an average over those trajectories leading to molecules in order to obtain the mean energy loss of the transmitted molecules. To speed up calculations we found it appropriate to replace $\phi^{(w)}$ by the approximation due to Vager and Gemmell [14]

$$\phi^{(w)}(z,\rho) = \frac{2Z}{\lambda_s} \sin(z/\lambda_s) K_0(\sqrt{\rho^2 + v^{-2}}/\lambda_s) \times \exp(z\gamma/2v_0)\Theta(-z) , \tag{10}$$

where (z,ρ) represent the coordinates parallel and perpendicular to the beam, respectively. Further, Z is the atomic number of the leading nucleus, γ describes the damping of the wake, $\lambda_s = v_0/\omega_p$ (where ω_p is the plasmon frequency), K_0 is the Bessel function of second kind and zeroth order, and $\Theta(x)$ is the unit step function.

Analogously, we calculated the stopping ratio [cf. Eq. (8)] using the stopping power from tables in Ref. [15] and the energy-loss straggling is simulated as a Gaussian-deviate random force having zero mean and a standard deviation obtained from straggling data in Ref. [16].

III. RESULTS AND DISCUSSION

A. Mean energy loss

Figure 1 shows the stopping ratio of H on C and Al foils as a function of the ion velocity. Open symbols are experimental data from Refs. [2,3,9]; full symbols are the results of our MC calculations. Although our MC calculations follow the experimental results fairly well, it must be noted that they appear approximately 10% above the experiments. For comparison, we plotted on the same figure the stopping ratio obtained by taking the average over all nuclei in the initial distribution. As one can easily imagine, this curve constitutes the "zero" dwell time limit, i.e., when all molecules can survive irrespective of initial orientation and/or separation. For a given foil, one would expect to have this stopping ratio at large bombarding energies. All the points shown in Fig. 1, however, are far from such a limit. The smallest dwell time of them all, at $v_0 = 1$, for which the dwell time is 2 fsec, corresponds to that of Fox and co-workers [9].

Deviations from the curve above indicate that the population of transmitted molecules differs from that in the initial distribution. Figure 2 shows the stopping ratio as a function of the ion velocity calculated for several internuclear orientations as well as separations of the nuclei in the molecule. A comparison of such results with experiments, shown as a dot-dashed line on the same figure, suggests that for velocities greater than approximately 1.5 a.u. transmission proceeds mainly from nuclei having a large internuclear separation; at low velocities transmitted clusters are from among the H_2^+ whose axes are parallel to the beam direction.

To further investigate this subject, we plotted on Figs. 3(a)–3(c) the relative position of the nuclei for 100 trajectories that yielded molecules. For reference purposes Fig. 3(a) shows the initial position of the nuclei. Figure

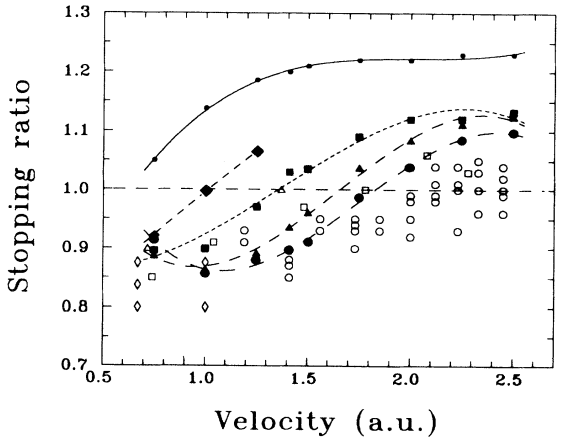

FIG. 1. Stopping ratio. Experiments appear as open symbols for 220 Å C, Ref. [3] (circles); 150 Å C, Ref. [2] (triangles); and 10–50 Å C, Ref. [9] (diamonds). Full symbols are our Monte Carlo calculations for 220 Å C (circles), 50 Å C (diamonds), and 200 Å Al (squares) targets, respectively. The dots represent the stopping ratio obtained for the clusters in the initial distribution and the curve is to guide the eye.

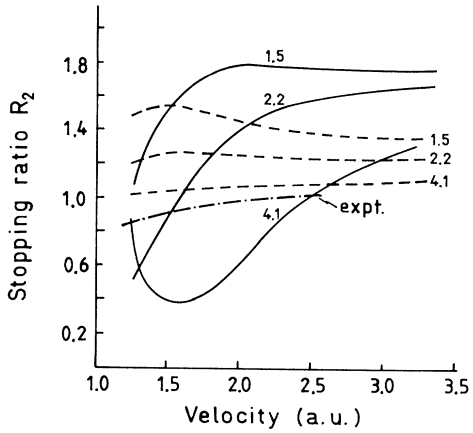

FIG. 2. Stopping ratio as a function of bombarding velocity for fixed orientation and separation of the nuclei in the molecule, respectively. Labels indicate separation in a.u. Orientations of the internuclear axis are parallel to the beam (continuous lines) and perpendicular to the beam (dashed lines), respectively. The stopping of the cluster was calculated using the wake potential in Ref. [14], whereas those of protons are from tables in Ref. [15].

3(b) displays the trajectories after 4 fsec. There the nuclei are seen slightly displaced from their initial position and those at small separations appear to be severely excluded from becoming molecules. With an increase of dwell time [cf. Fig. 3(c)] the trajectories become larger and the nuclei appear to have been driven towards symmetric positions along the beam direction. These results appear to be in remarkable agreement with our previous "guess" about orientations and separations of the nuclei in the transmitted molecules.

Looking at the potential energy in Fig. 4, it is evident that clusters with small separations can hardly yield molecules because they will *explode* under the effects of the Coulomb repulsion. Analogously, one can see the potential *well* that appears along the z axis, in coincidence with the sites where the nuclei were seen "trapped" at a longer dwell time. We did not go further on this matter since a detailed discussion about the motion of the nuclei in the wake potential will be presented in a forthcoming paper [17]. It is important to mention that other wake potentials were also used to calculate R_2 too. The results, however, do not differ from those shown in Fig. 1. The so-called local dielectric approximation yielded a slightly small vicinage effect, particularly at low velocities. These results, however, were not at all unexpected, since such a dielectric function is known to give a small wake potential. Using a dielectric function that includes plasmon dispersion [see Eqs. (20) and (21) in Ref. [10]], the results are close to those obtained with the Vager-Gemmell approximation [cf. Eq. (10)]. Differences, on the other hand, become more pronounced at low energies where the wake may not be very reliable.

B. Foil inhomogeneity

As mentioned at the beginning of this paper, the vicinage effect, i.e., deviation from unity of the stopping ra-

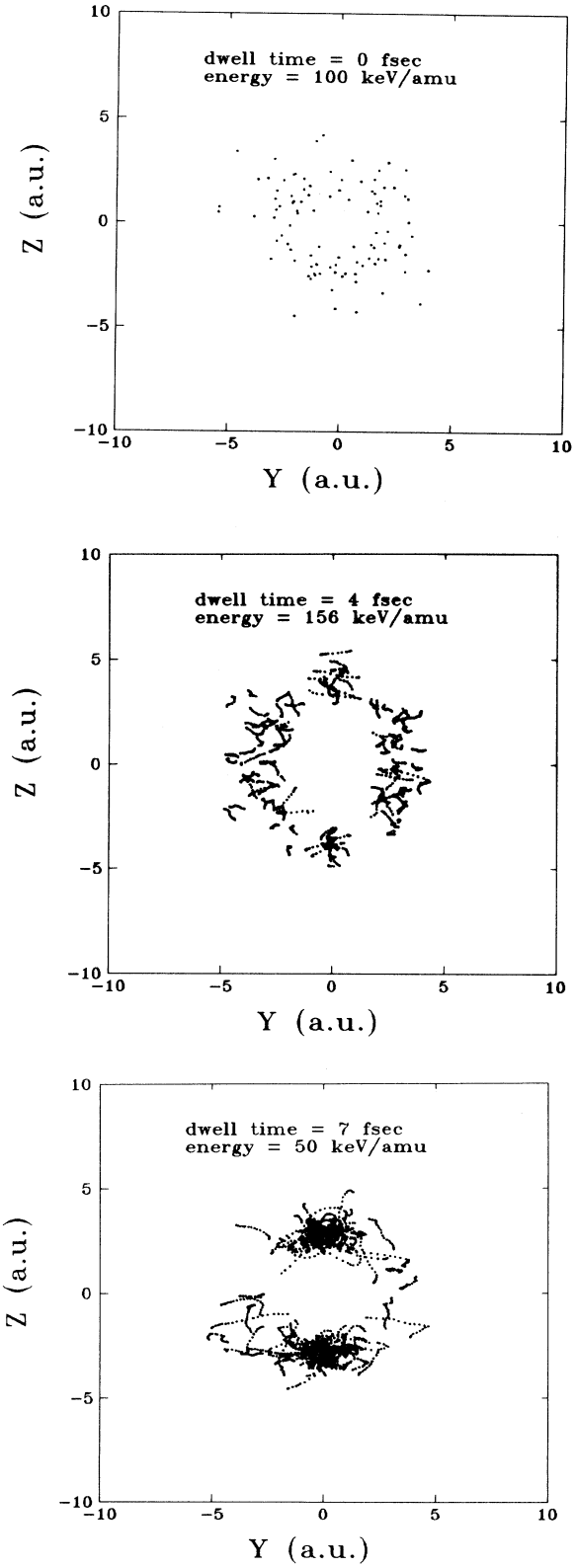

FIG. 3. Relative position of the nuclei for 100 trajectories that led to molecules: (a) initial distribution, i.e., 0 fsec; (b) 156 keV/amu and 4 fsec dwell time; and (c) 50 keV/amu and 7.1 fsec dwell time. In (b) and (c) the points indicate the relative position every 0.33 fsec.

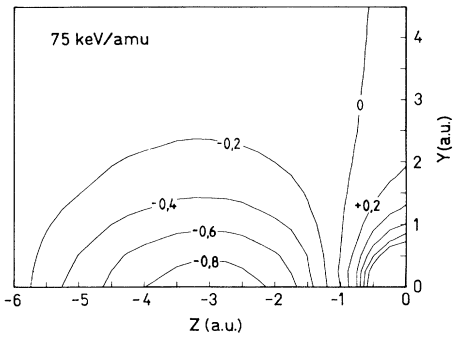

FIG. 4. Potential energy (in atomic units) for 75 keV/amu H_2^+ as a function of relative position. The oscillatory part of the wake potential is obtained from approximate expression in Ref. [14]. The screened Coulomb is approximated by a function of the form $r^{-1}\exp(-r/\lambda_s)$ [see the paragraph following Eq. (10)]. Note that the contour lines for $E_{\rm pot}>1$ a.u. are suppressed.

tio, can be also produced by nonuniformities of the foil. In fact, as discussed in Ref. [13], even if there are no vicinage effects, the mean energy loss of transmitted clusters will be smaller than that of the protons at the same velocity, provided the foil contains thickness inhomogeneities. To estimate such an effect, let us assume that the foil contains a distribution of thickness $P(\Delta x)$ and $Y(\Delta x)$ represents the transmission probability for the H_2^+ ions as a function of the foil thickness. To the transmitted molecules, the mean thickness of the foil thus becomes

$$\langle \Delta x \rangle_{\rm mol}=\frac{\int P(\Delta x)Y(\Delta x)\Delta x\, d\Delta x}{\int P(\Delta x)Y(\Delta x)d\Delta x} . \tag{11}$$

Assuming that Y varies with thickness slower than P, one can thus expand Y in a power series of Δx around the mean foil thickness, i.e., $\langle \Delta x \rangle$. Hence Eq. (11) yields

$$\langle \Delta x \rangle_{\rm mol}\approx\langle \Delta x \rangle(1+\nu\rho^2) , \tag{12}$$

where

$$\langle \Delta x \rangle=\int P(\Delta x)\Delta x\, d\Delta x , \tag{13}$$

ρ is the *roughness coefficient* [13] defined as

$$\rho^2=\frac{1}{\langle \Delta x \rangle^2}\int P(\Delta x)(\Delta x-\langle \Delta x \rangle)^2 d\Delta x , \tag{14}$$

and

$$\nu=\left.\frac{\partial \ln Y}{\partial \ln \Delta x}\right|_{\Delta x=\langle \Delta x \rangle} . \tag{15}$$

Therefore, the stopping cross section for the molecular ion $S_{\rm mol}$ relates to the mean energy loss for transmitted molecules as [18]

$$\Delta E(H_2^+,H_2^+)=NS_{\rm mol}\langle \Delta x \rangle_{\rm mol} , \tag{16}$$

according to Eq. (12),

$$\Delta E(H_2^+,H_2^+)\approx NS_{\rm mol}\langle \Delta x \rangle(1+\nu\rho^2) . \tag{17}$$

Hence the stopping ratio in (8) can be written as

$$R_2=\frac{S_{\rm mol}}{2S_{\rm proton}}(1+\nu\rho^2) \tag{18}$$

or

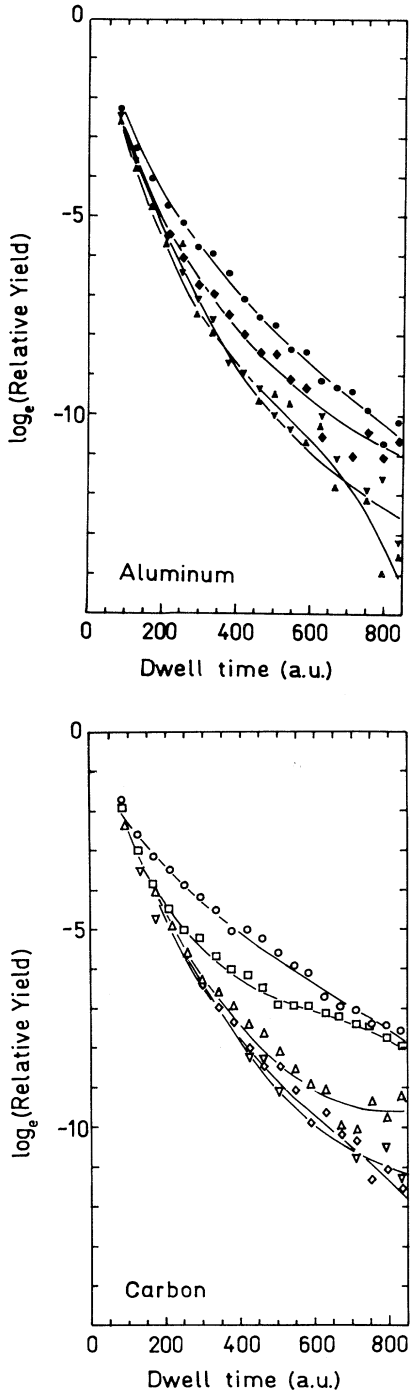

FIG. 5. Calculated relative transmission yields for H_2^+ on carbon and aluminum foils. (a) Carbon targets: 50 (circles); 100 (squares); 200 (up-pointed triangles); 300 (diamonds), and 1000 (down-pointed triangles) keV/amu, respectively. (b) Aluminum targets: 25 (full circles); 50 (squares); 100 (up-pointed triangles), and 300 (down-pointed triangles) keV/amu, respectively.

$$R_2^{(\text{expt})} = R_2^{(\text{theor})}(1+\nu\rho^2) , \tag{19}$$

with $R_2^{(\text{theor})} = S_{\text{mol}}/2S_{\text{proton}}$. Consequently, if one assumes that $S_{\text{mol}} = 2S_{\text{proton}}$ or $R_2^{(\text{theor})} = 1$, we thus have $R_2^{(\text{expt})} = 1+\nu\rho^2$. That is to say, $R_2^{(\text{expt})} \neq 1$ despite the fact that no correlation in the stopping was assumed. We can readily conclude that the stopping ratio $R_2^{(\text{expt})}$ will lead to $S_{\text{mol}}/2S_{\text{proton}}$ if and only if the foil contains no roughness. Observe also that as ν is presumably always less than zero, then $R_2^{(\text{expt})} \leq R_2^{(\text{theor})}$, where the equal sign holds only for homogeneous foils.

From our own calculations of the transmission yield for H_2^+ on C and Al, shown in Figs. 5(a) and 5(b) [19], we have obtained the corresponding ν's [cf. Eq. (15)]. The results are plotted in Figs. 6(a) and 6(b) as a function of the ion velocity v and different foil thicknesses. There, we can readily see that ν increases (in an absolute sense) with increasing the foil thickness. Furthermore, ν becomes nearly a constant for $v > 1.5$. a.u., whereas it appears to be a rapidly varying function of the velocity for $v < 1.5$ a.u. Within this region ν is seen to fall to large negative values with decreasing v. It should also be noted that the results for aluminum appear to be greater (in an absolute sense) than those of carbon at same velocity and thickness, respectively.

According to Eq. (19) and the results in Fig. 5, we can conceivably expect that the importance of inhomogeneities will be greater for aluminum than for carbon foils. Analogously, it appears that using thicker foils does not necessarily warrant a smaller influence of the foil roughness unless of course, a small roughness coefficient was attained. Notice also that if velocity is lowered, a larger influence of the inhomogeneity is expected. This is a somewhat unfortunate result since at low velocities the vicinage effect shows such a distinctive less-than-unity stopping ratio, which has attracted the attention of several workers in the field [2,8].

As an application of our results, we have calculated the roughness coefficients for the foils used in Refs. [3,2,9]. To this end we minimize the root-mean-square (rms) error between expression (19) and the R_2's from experiments, where the $S_{\text{mol}}/2S_{\text{proton}}$'s are obtained from MC calculations as explained in Sec. III A. Furthermore, we have assumed that the roughness coefficients corresponding to the same element and the same laboratory are all equal. In Table I one can see the rms errors between our calculations and experiments assuming no roughness, i.e., $\rho=0$, and those resulting after the minimization. Analogously, the fifth column contains the roughness

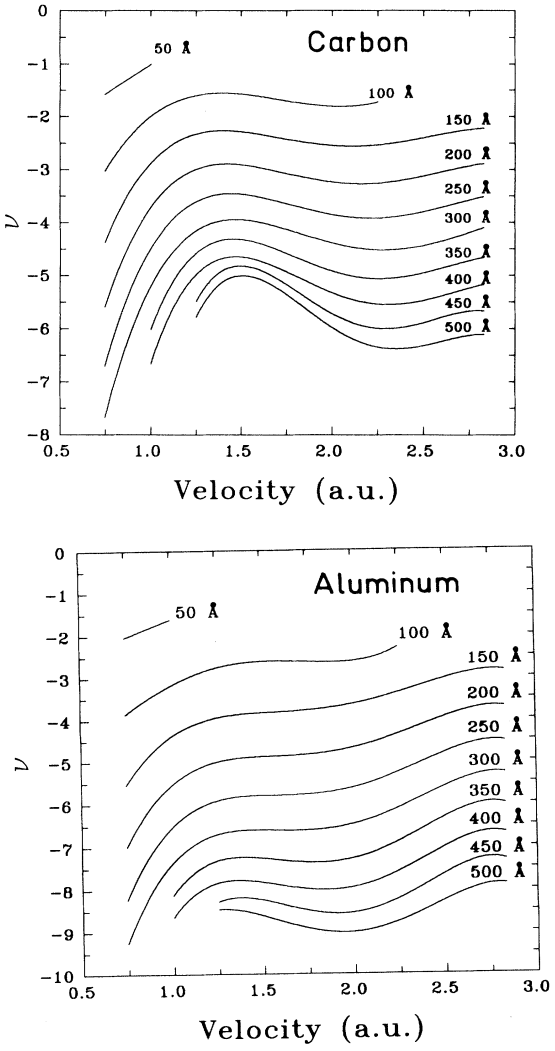

FIG. 6. ν defined in Eq. (15) for the transmission yields in (a) carbon and (b) aluminum, respectively.

coefficients that produce the best fit between experiments and our calculations. Observe that the two points for carbon in Ref. [2] cannot be included in our minimizing procedure because these data are greater than our calculations; thus, as ν is *always* less than zero [see Fig. 6(a)], the minimizing procedure will necessarily lead to $\rho=0$.

Notice that rms errors can be reduced by approximately a factor of 2 after minimization. Theoretical results,

TABLE I. Roughness coefficients obtained by fitting our calculations with experimental results according to Eq. (19).

Target	Foil thickness (Å)	rms error ($\rho=0$)	rms error ($\rho_{\min}$)	$\rho_{\min}$	Source
C	220	0.077	0.043	0.134	[3]
C	50	0.16	0.076	0.253	[9]
C	150	0.054			[2]
Al	200	0.070	0.033	0.109	[2]

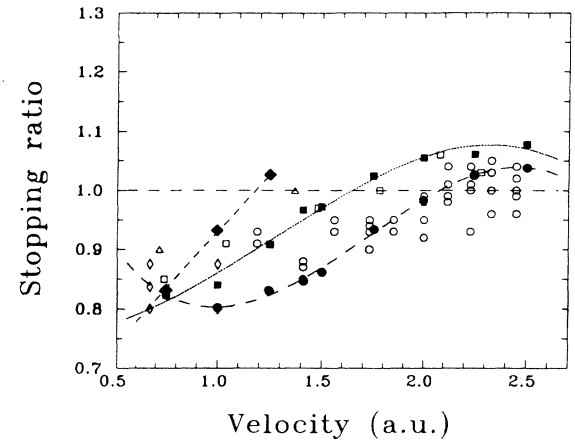

FIG. 7. Stopping ratio. Experiments are the same as those in Fig. 1, but Monte Carlo results (filled symbols) were previously multiplied by $(1+\nu\rho_{\min}^2)$ with $\rho_{\min}$ from Table I.

however, cannot perfectly fit experiments since data appear to have an intrinsic statistical uncertainty of the order of 5%. Nevertheless, it is worth noticing that the roughness coefficients calculated according to our minimizing procedure and using our MC calculations do agree reasonably well with our previous estimates in Ref. [13]. Finally, in Fig. 7 we show again the stopping ratio, though this time the theoretical results were previously multiplied by $(1+\nu\rho_{\min}^2)$ in order to have them "corrected" for inhomogeneities. As one can see, the agreement between experiment and theory appears to have improved notably.

IV. CONCLUDING REMARKS

We have carried out Monte Carlo calculations of the mean energy loss for H_2^+ transmitted through thin carbon and aluminum foils. Using the wake potential in Ref. [14], we obtained a reasonable agreement with experimental results. The velocity dependence of the vicinage effect was found to be a consequence of the wake forces and the way transmission "selects" from among the clusters depending on the nuclei trajectories. We found a small, though systematic, deviation between calculations and experimental data, which is attributed to the inhomogeneities of the target thickness. By means of a procedure given in [13], we calculated the roughness coefficients of the target used in the experiments. The results are surprisingly consistent with previous estimations [13], but the uncertainties accompanying both theory and experimental data do not allow one to be confident about these results. Altogether, after calculating the transmission yields of H_2^+ for low bombarding energies—data that were not available at the time we published Ref. [13]—we found again that the influence of thickness inhomogeneity becomes more pronounced with a decrease in the ion velocities. Hence, since at low energies the vicinage effect is expected to produce stopping ratios smaller than unity and the inhomogeneity, in turn, reduces the stopping ratio, one might mistake one effect for the other. A more complete study of the passage of H_2^+ ions through thin films is currently being completed [17]. The energy-loss and angle spectra of the transmitted molecular ions appear quite revealing since they show aspects of correlation between charge particles that cannot be seen on integrated quantities such as transmission yields or the mean energy loss.

ACKNOWLEDGMENTS

One of us (M.M.J.) would like to thank the A. von Humboldt Foundation for providing financial support during an earlier stage of this work. This work was supported in part by Gobierno Autónomo de Canarias, through the Conserjería de Educación, Cultura y Deportes.

[1] W. Brandt, A. Ratkovsky, and R. Ritchie, Phys. Rev. Lett. **33**, 1325 (1974); **35**, 130 (1975).
[2] J. C. Eckardt, G. H. Lantschner, N. R. Arista, and R. A. Baragiola, J. Phys. C **11**, L851 (1978).
[3] R. Laubert, IEEE Trans. Nucl. Sci. **NS-26**, 1020 (1979).
[4] W. Brandt and R. Ritchie, Nucl. Instrum. Methods **132**, 43 (1976).
[5] D. S. Gemmel, J. Remillieux, J. C. Poizat, M. E. Gailliard, R. E. Holland, and Z. Vager, Phys. Rev. Lett. **34**, 1420 (1975); Nucl. Instrum. Methods **132**, 61 (1976).
[6] M. F. Steuer, D. S. Gemmel, E. P. Kanter, and G. J. Zabransky, IEEE Trans. Nucl. Sci. **NS-30**, 1069 (1983).
[7] J. W. Tape, W. M. Gibson, J. Remillieux, R. Laubert, and H. E. Wegner, Nucl. Instrum. Methods **132**, 75 (1976).
[8] J. Steinbeck, M. W. Lucas, J. Kemmler, and K.-O. Groeneveld, Nucl. Instrum. Methods Phys. Res. Sect. B **48**, 51 (1990).
[9] T. R. Fox, K. Lamb, and R. Levi-Seti, Nucl. Instrum. Methods **194**, 285 (1982); see also R. Levi-Seti, K. Lamb, and T. R. Fox, *ibid.*, **194**, 281 (1982).
[10] P. Echenique, R. Ritchie, and W. Brandt, Phys. Rev. B **20**, 2567 (1979).
[11] P. Echenique, F. Flores, and R. H. Ritchie, Solid State Phys. **43**, 229 (1990).
[12] M. M. Jakas and N. E. Capuj, Nucl. Instrum. Methods Phys. Res. Sect. B **93**, 14 (1994).
[13] M. M. Jakas and N. E. Capuj, Phys. Rev. A **40**, 7369 (1989).
[14] Z. Vager and D. Gemmell, Phys. Rev. Lett. **37**, 1352 (1976).
[15] J. F. Ziegler, J. P. Biersack, and U. Littmark, *The Stopping and Ranges of Ions in Matter* (Pergamon, New York, 1985), Vol. I.
[16] W. K. Chu, IBM Technical Report No. TR22, 1974 (unpublished); Phys. Rev. A **13**, 2057 (1976).
[17] M. M. Jakas and N. E. Capuj, J. Phys. C (to be published).
[18] Note that in writing Eq. (16), $S_{\rm mol}$ was not included within the average bracket. This is certainly an approximation since $S_{\rm mol}$ depends on the nuclei trajectory. The error will be negligible, however, provided $S_{\rm mol}$ and Δx are found to be weakly correlated along different trajectories.
[19] Although we use the *absolute* transmissition probability in Eq. (11), i.e., Y, one can replace Y by the relative yield defined as $Y/2\phi_0$, where ϕ_0 is the neutral fraction of protons at the same velocity. Note that since ϕ_0 does not depend on the thickness, it will canceled out.