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# Monte Carlo calculation of the energy-loss spectra for fast $H_2^+$ molecular ions transmitted through thin foils

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**Abstract.** We report calculations of the energy-loss spectra for 25–300 keV amu$^{-1}$ $H_2^+$ ions transmitted through carbon and aluminium foils. At 25 keV amu$^{-1}$ the mean energy-loss of transmitted $H_2^+$ ions is approximately 15% smaller than that of two protons at the same velocity, whereas at 300 keV amu$^{-1}$ it becomes 10% greater. When compared to those of two uncorrelated protons, the width of the energy-loss distribution appears to increase with a decrease of the bombarding energy, becoming 3–5 times larger at 25 keV amu$^{-1}$ for molecules transmitted through aluminium and carbon foils, respectively.

## 1. Introduction

It is well known that the mean energy-loss of molecular ions differs from those of the constituent nuclei at the same velocity. This phenomenon, known as the *vicinage* effect on the stopping, has been confirmed by a number of experiments (Brandt *et al* 1974, 1975, Brandt and Ritchie 1976, Gemmell *et al* 1975, 1976, Tape *et al* 1976, Eckardt *et al* 1978, Laubert 1979, Fox *et al* 1982, Levi-Seti *et al* 1982, Steuer *et al* 1983, Steinback *et al* 1990). On the theoretical side, however, while a great deal of effort has gone into the calculation of the stopping power and energy-loss straggling for *fixed* nuclei in the cluster (Echenique *et al* 1979, 1990), a complete study of the passage of $H_2^+$ ions through thin films, including energy loss, is lacking. However, there seems to be no doubt that a fast charged particle induces a wake of electron density fluctuation trailing the charge, so that a second charge moving within that wake is subjected to a modified stopping. This explains the observed deviation of the stopping of clusters from that of the uncorrelated nuclei; more importantly, such deviations give us the unique possibility of investigating the spatial and time extension of such a polarization wake.

In order to calculate accurately the stopping of transmitted $H_2^+$ ions one needs the trajectories of the nuclei that become bound molecules. Unfortunately, the passage of correlated atomic nuclei through thin films poses a very complicated computational problem. Screened Coulomb repulsion, inelastic slowing down, multiple-scattering by target atoms and wake forces all act (simultaneously) upon the nuclei during penetration. The motion of a particle subject to forces having different spatial and time characteristics as those mentioned above is not amenable to exact analytical solution; the situation does not seem to be much better using numerical simulations. The main difficulty here stems from the fact that transmission yields are often so small that the use of conventional (direct) Monte Carlo methods (MC) would require excessively large computation times.

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Based on two MC techniques (*killing* and *splitting*) we developed an MC code that has been proven to deal with the transmission of $H_2^+$ ions within fairly small computation times (Jakas *et al* 1994). Using such an MC program, in this paper we report for the first time Monte Carlo calculations of the energy-loss spectra for $H_2^+$ transmitted through carbon and aluminium foils. We begin in section 2 by giving the assumptions used to calculate the energy loss for the two protons in the cluster. The details of our MC code have been omitted, since they have been published elsewhere (Jakas *et al* 1994). The results are presented in section 3. First, we compare the calculated mean energy-loss with available experimental data. Second, our results are qualitatively explained by investigating the relative motion of the nuclei in the field of the wake potential. Third, calculations of the energy-loss spectra for the transmitted molecules are presented, and the full width at half maximum (FWHM) relative to that of uncorrelated protons are displayed as a function of the bombarding energy. Section 4 concludes this paper with a brief summary.

## 2. Theory

First we assume that, at the point of penetration, the nuclei dissociate. The centre-of-mass (COM) of the H fragments is thus subjected to a force

$$\boldsymbol{F}_{\mathrm{COM}} = \boldsymbol{F}_1 + \boldsymbol{F}_2 \tag{1}$$

where $\boldsymbol{F}_i$ is the force acting upon the $i$th nucleus, with $i = 1, 2$. The two forces (1) can be separated into the following components†

$$\boldsymbol{F}_i = \boldsymbol{F}_i^{(\mathrm{w})} + \boldsymbol{F}_i^{(\mathrm{C})} + \boldsymbol{F}_i^{(\mathrm{stp})} \qquad i = 1, 2 \tag{2}$$

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

Accordingly, the energy loss after a dwell time $\tau$ in the foil becomes

$$\Delta E_{\mathrm{COM}}(\tau) = -\int_0^{\tau} \mathrm{d}t\, \boldsymbol{v} \cdot \boldsymbol{F}_{\mathrm{COM}}. \tag{3}$$

Since the Coulomb forces are unable to modify the energy of the centre-of-mass, one may thus write

$$\Delta E_{\mathrm{COM}}(\tau) = \Delta E_1^{(\mathrm{stp})}(\tau) + \Delta E_2^{(\mathrm{stp})}(\tau) + \Delta E^{(\mathrm{w})}(\tau) \tag{4}$$

where $\Delta E_{1(2)}^{(\mathrm{stp})}(\tau)$ denotes the *uncorrelated* energy loss of the nucleus 1(2), respectively. Furthermore, given that the wake force acts only upon the trailing ion in the cluster we can write

$$\Delta E^{(\mathrm{w})}(\tau) = -\int_0^{\tau} \mathrm{d}t\, \boldsymbol{v} \cdot \boldsymbol{F}_{\mathrm{trail}}^{(\mathrm{w})}. \tag{5}$$

† Although our Monte Carlo calculations include the impulsive forces coming from the elastic scattering by target atoms, they are purposely omitted in (2). That is an acceptable approximation since, as far as the energy of the COM is concerned, such impulsive forces act indirectly through the wake force, i.e. by modifying the nuclei trajectory within the wake potential.

Finally, as the velocity of the protons during the passage does not change for practical purposes, one can replace $v$ by the initial velocity $v_0$. Hence, assuming that the $z$ axis is along the beam direction, we have

$$\Delta E^{(w)}(\tau) = v_0|e| \int_0^\tau dt \frac{\partial \phi^{(w)}}{\partial z} \tag{6}$$

where $\phi^{(w)}$ is the 'oscillatory' part of the wake potential and $e$ is the elementary charge. Here, in order to speed up calculations, we find it convenient to replace $\phi^{(w)}$ by the approximation given by Vager *et al* (1976), which in atomic units can be written as

$$\phi^{(w)}(z, \rho) = \frac{2Z}{\lambda_s} \sin(z/\lambda_s) K_0\left[(\rho^2 + v_0^{-2})^{1/2}/\lambda_s\right] \exp(z\gamma/2v_0)\Theta(-z) \tag{7}$$

where $\rho = (x^2 + y^2)^{1/2}$. Furthermore, $Z$ is the atomic number of the leading nucleus, $\gamma$ describes the damping of the wake, $\lambda_s = v_0/\omega_p$ where $\omega_p$ is the plasmon frequency, $K_0$ is a Bessel function of the second kind and zeroth order, and $\Theta(x)$ is the unit step function.

As was stated following (2), the *uncorrelated* energy-loss of the two protons is simulated by changing the velocity of the nuclei along the beam direction after each time step. These changes are realized by adding one non-stochastic velocity, representing the mean energy-loss, plus a Gaussian-distributed velocity which accounts for the energy-loss straggling; here the standard deviation of the distribution is obtained from the straggling tables given by Chu (1974). Therefore, one has

$$\Delta E_{1(2)}^{(stp)}(\tau) = v_0 \tau N S_p + \Delta E_{1(2)}^{(strg)}(\tau) \tag{8}$$

where $NS_p$ and $\Delta E_{1(2)}^{(strg)}(\tau)$ denote the stopping power and the energy-loss straggling, respectively.

Since the non-stochastic part of the stopping is the same for the two nuclei, it can be disregarded or, equivalently, the energy loss can be measured with respect to that of the two uncorrelated protons as

$$\widetilde{\Delta E}_{COM}(\tau) = \Delta E_{COM} - 2v_0 \tau N S_p = \Delta E_1^{(strg)}(\tau) + \Delta E_2^{(strg)}(\tau) + \Delta E^{(w)}(\tau). \tag{9}$$

Taking an average over the trajectories that produced molecules one obtains

$$\langle \Delta E_{COM} \rangle_{mol}(\tau) = 2v_0 \tau N S_p + \langle \Delta E^{(w)} \rangle_{mol}(\tau). \tag{10}$$

Observe that in writing (10) we have made use of the fact that $\langle \Delta E_{1(2)}^{(strg)} \rangle_{mol}(\tau) = 0$.

Following Laubert (1979), we introduce the stopping ratio $R_2$, defined as the ratio between the mean energy-loss of transmitted molecules to twice that of protons at the same velocity. According to equation (10) one has

$$R_2 = 1 + \frac{\langle \Delta E^{(w)} \rangle_{mol}(\tau)}{2v_0 \tau N S_p}. \tag{11}$$

The term $R_2$ is the relevant quantity here since, as equation (11) shows, a deviation of $R_2$ from unity implies that the wake forces are non-zero or, alternatively, that the stopping of the clusters differs from that of two uncorrelated protons at the same velocity.

### *2.1. The Monte Carlo program*

Although our Monte Carlo code has been described elsewhere (see Jakas *et al* 1994), for the sake of completeness we mention that the simulation begins by selecting the relative position of the nuclei. Afterwards, the motion of the nuclei are calculated by dividing the dwell time into a number of time steps. In each time step the nuclei undergo both scattering with a target atom and energy loss straggling. Furthermore, the nuclei are subjected to their own Coulomb repulsion, the wake force, and a constant force representing the stopping power so that, between time steps, the motion of the nuclei is calculated by means of a size-step adaptive integrator. Finally, at the exit of the foil, the nuclei may capture an electron into a binding state. Thus, provided such a capture has occurred, there will be a $H_2^+$ ion if the total energy of the nuclei is less than or equal to zero, i.e. $U_g(r) + E_k \leqslant 0$, where $U_g$ is the potential energy in the binding state, $r$ is the separation between the nuclei, and $E_k$ is the relative kinetic energy.

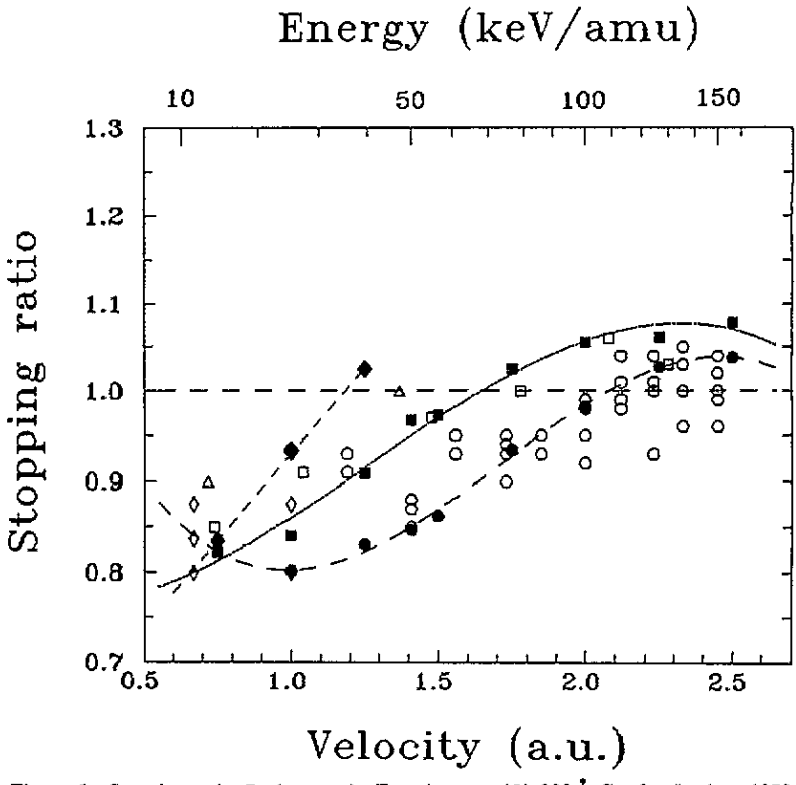


**Figure 1.** Stopping ratio $R_2$ (see text). Experiments: (○) 220 Å C, after Laubert 1979; (△) 150 Å C, after Eckardt *et al* 1978; (◇) 10–50 Å C, after Fox *et al* 1982. Monte Carlo calculations: (●) 220 Å C; (♦) 50 Å C; (■) 200 Å Al.

## 3. Results and discussion

Figure 1 shows the stopping ratio for $H_2^+$ transmitted through C and Al foils, from Laubert (1979), Eckardt *et al* (1978), Fox *et al* (1982) and Levi-Seti *et al* (1982). As

one can see, at energies lower than approximately 70 keV amu$^{-1}$, $R_2$ is smaller than one, whereas at higher energies $R_2$ becomes slightly greater than one. The $R_2$ calculated using our MC are plotted in the same figure. In this case we use (11), with $\langle \Delta E^{(w)} \rangle_{mol}(\tau)$ obtained from simulations and the stopping power $NS_p$ from tables given by Ziegler *et al* (1985). The damping coefficient $\gamma$ was set to 0.27 and 0.35 au for carbon and aluminium, respectively. As one can see, the agreement between calculations and experiments is remarkable.

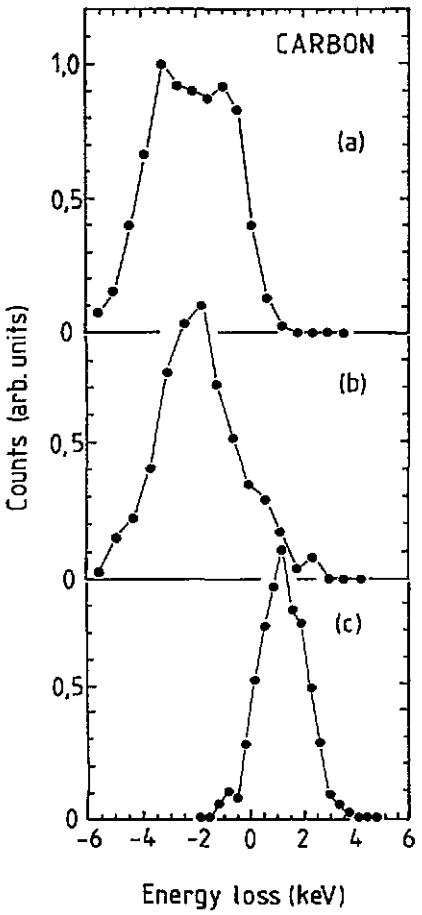


**Figure 2.** Monte Carlo calculations of the energy-loss spectra for $H_2^+$ ions transmitted through carbon foils: (*a*) 25, (*b*) 50 and (*c*) 300 keV amu$^{-1}$, respectively.

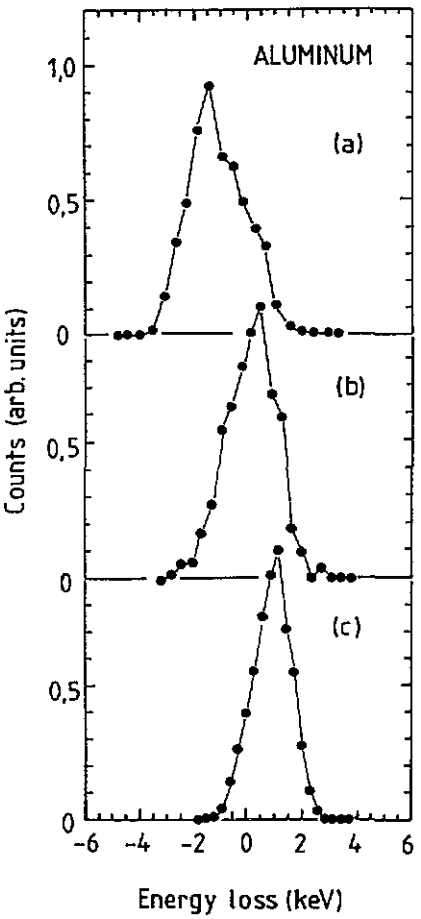


**Figure 3.** Monte Carlo calculations of the energy-loss spectra for $H_2^+$ ions transmitted through aluminium foils: (*a*) 25, (*b*) 50 and (*c*) 300 keV amu$^{-1}$, respectively.

To further investigate this matter we show in figures 2 and 3 the energy-loss spectra for the transmitted $H_2^+$ ions. Notice that the energy loss displayed in these figures is measured with respect to that of uncorrelated protons, i.e. $\widetilde{\Delta E}_{COM}$. Therefore, a shift towards positive values is associated with $R_2 > 1$ in the same manner as $\widetilde{\Delta E}_{COM} < 0$ is related to $R_2 < 1$.

One can see in figures 2 and 3 that, in agreement with the experiments shown in figure 1, at large bombarding energies the energy-loss distribution appears shifted towards positive values. At lower energies, however, the distribution becomes not only displaced towards

negative values but also notably wider. Figure 4 shows the FWHM of the molecular spectra relative to those that one would expect for two uncorrelated protons†. One can see that at low bombarding velocity the FWHM becomes between three to five times wider than that of independent protons. Moreover, going from high to low energies, the FWHM is seen to start increasing at approximately the same velocity where the stopping ratio becomes equal to unity in figure 1.

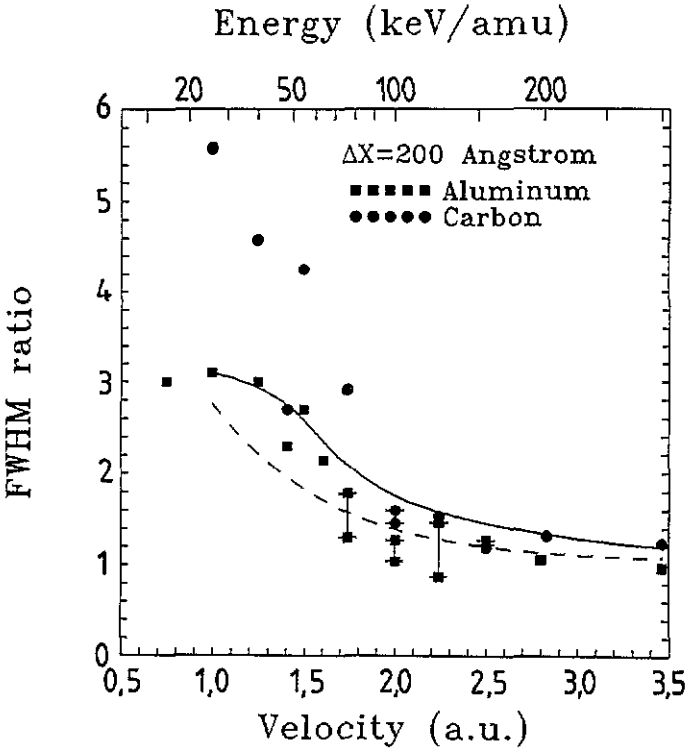


**Figure 4.** FWHM of the energy-loss distribution relative to that of two uncorrelated protons at the same velocity. Calculations are for 200 Å-thick carbon and aluminium foils. Curves indicate the 'static' limit, i.e. assuming that the nuclei remain fixed during passage through the foil. The full curve is for carbon and the broken curve for aluminium.

Previous results can be understood by looking at the slope of the wake potential displayed in figures 5 and 6. One must recall that such a potential is produced by the leading proton in the cluster, which is assumed to be exactly at the origin. Similarly, the screened Coulomb repulsion is ignored since it cannot alter the velocity of the centre-of-mass. Finally, the thick curves show (approximately) the minimum and maximum distances between the nuclei in the initial distribution.

Looking at the slope of the wake potential, one can see that at high energy all the nuclei are located within a region where the wake force opposes the ion velocity. This explains

† Despite the fact that the energy-loss straggling entering our simulations may not be accurate, the results in figure 4 are self-consistent in the sense that the straggling assumed for the independent protons is the same as that used for the molecules in the simulations.

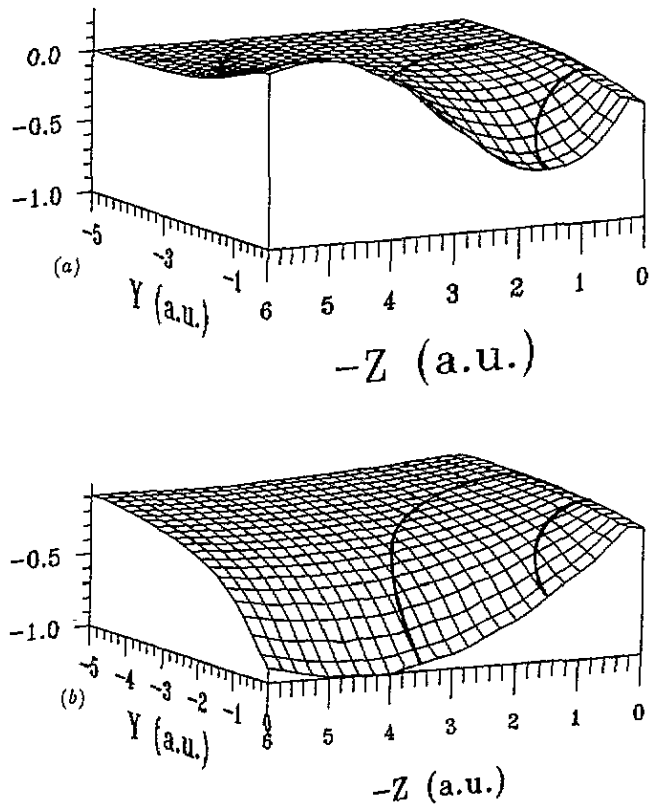


**Figure 5.** Wake potential for proton in carbon. (*a*) 25 and (*b*) 200 keV amu$^{-1}$, respectively. A proton is assumed to be at the origin and moving parallel to the $z$ direction, and the potential is evaluated on the $y$–$z$ plane; see below equation (10).

both the shift of the energy-loss distribution towards positive values in figures 2 and 3 and the enlarged mean energy-loss observed in figure 1. Furthermore, as the wake forces appear not to vary very much within the initial internuclear distribution, no additional straggling is expected in this case.

At low energy the situation is different. As the minimum of the wake potential is closer to the origin, clusters with large internuclear separations now reach the farthest side of the well. The wake forces acting on these clusters will be directed towards the ion velocity, while for those at small separations the wake forces remain opposite to the beam direction. This implies that for one part of the incoming molecules the stopping is reduced while for the others, i.e. those at small separations, it is increased (see figures 2 and 3). Although the net effect of these two contributions cannot be intuitively predicted, it is clear that because of that feature the energy-loss distribution should be wider.

According to our previous discussion, it is apparent that the broadening of the energy-loss distribution stems from a disparity of the wake forces. Owing to displacements of the nuclei with dwell time, the above arguments will now not be totally accurate, particularly at low bombarding energies. The extent to which the motion of the nuclei contributes to the energy-loss broadening can be seen in figure 4. The lines in this figure display the broadening that one would expect from the wake forces (in the approximation given by (7)) assuming that the nuclei are fixed, i.e. the 'static' limit. For bombarding energies greater

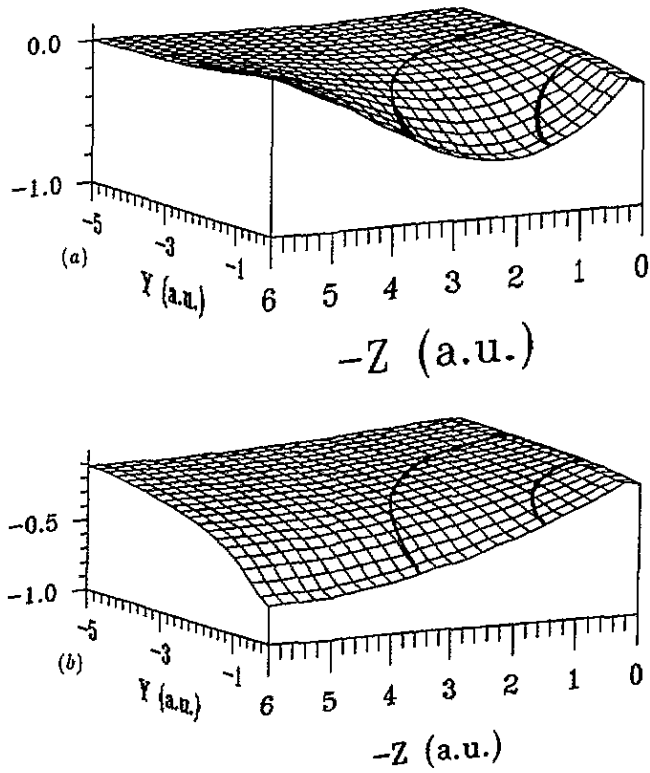


**Figure 6.** Wake potential for proton in aluminium. (*a*) 25 and (*b*) 200 keV amu$^{-1}$, respectively. A proton is assumed to be at the origin and moving parallel to the $z$ direction, and the potential is evaluated on the $y$–$z$ plane; see below equation (10).

than, say, 70 keV amu$^{-1}$ it appears that displacements of the nuclei can be ignored. For lower energies, however, and particularly for carbon, the results of our MC calculations are greater than those in the static limit. This indicates that, the nuclei having been displaced, they have passed through less homogeneous regions in the wake potential, thus producing an additional broadening.

## 4. Concluding remarks

In summary, we have calculated the energy-loss spectra of $H_2^+$ transmitted through thin C and Al foils using Monte Carlo simulations. Our results agree with the experimental results of Laubert (1979), Eckardt *et al* (1978), Fox *et al* (1982) and Levi-Seti *et al* (1982). According to our calculations, the behaviour of the stopping ratio $R_2$ is linked to variations of the wake forces with the bombarding energy within the relevant internuclear distances. At high energy we found that the wake forces are relatively homogeneous and directed against the beam direction. This causes the energy-loss distribution to be shifted towards high energy-losses, but with no broadening. That is, the FWHM does not differ from that of two uncorrelated protons.

At lower energies the wake forces can be directed against or towards the beam direction, depending on whether the separation between the nuclei is small or large, respectively. As

a consequence, the energy-loss spectra become nearly split into two parts: one is shifted to small losses, and the other is shifted to high energy-losses, respectively. This leads to a reduction of the mean energy-loss and, most importantly, to an increase in the width of the energy-loss distribution. In this regard, the wake forces appear to have a much larger influence upon the width of the energy-loss distribution than they do upon the mean energy-loss. To the best of our knowledge, however, no experiment has so far been reported about this energy-loss broadening. As a final remark we would like to note that, for 200 Å-thick carbon or aluminium foils and bombarding energies lower than 70 keV amu$^{-1}$, the FWHM of the energy-loss distribution for transmitted $H_2^+$ ions would be several times larger than those of uncorrelated protons (ranging from three times larger for aluminium, up to five times larger for carbon foils, respectively).

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