The influence of the Coulomb explosion on the energy loss of H_2^+ and H_3^+ molecules channeling along the Si ⟨100⟩ direction

R.C. Fadanelli, P.L. Grande, M. Behar, J.F. Dias

Instituto de Física da Universidade Federal do Rio Grande do Sul, Avenida Bento Gonçalves 9500, C.P. 15051, CEP 91501 - 970, Porto Alegre, Brazil

G. Schiwietz

Hahn-Meitner-Institut, Abteilung SF4 Glienicker Strasse 100, D-14109 Berlin, Germany

Abstract

In this work we have measured the contribution of the Coulomb explosion to the electronic stopping power of molecular hydrogen ions (H_2^+ and H_3^+) channeling along the Si ⟨100⟩ direction. To this end, we have used a SIMOX target, consisting of a crystalline ⟨100⟩ Si with a buried layer of SiO_2. The measurements of the energy loss of H^+, H_2^+ and H_3^+ have been carried out using the standard channeling Rutherford Backscattering Spectrometry. The energy loss have been measured around the Si ⟨100⟩ channel with the same energy per nucleon (150 keV / a.m.u.) as a function of the tilt and azimuthal angles. The present results show the effect of Coulomb explosion, which enlarges the protons traversal energy and consequently the channeling energy loss. This heating effect due to H_3^+ ions is about two times larger than H_2^+ molecules and amounts to about 5% of the total stopping power.

Key words: channeling, energy loss, Coulomb explosion
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1. Introduction

Beams of ionic clusters are useful tools in both fundamental and applied researches, namely material science, plasma and nuclear physics. It is well established that the effects of a molecular beam clearly deviates from those related with its individual components. The first evidence of interference effects among the projectile constituents, namely the vicinage effect, was first reported by Brandt et al. [1] and it was further studied by several authors, e.g. [2,3]. A detailed review of the subject is given in ref. [4].

A swift molecular beam, when entering in a solid, shows a second effect. The projectile looses its bonding electrons in the target and its components undergo a molecular breakup process due to quasi-Coulomb repulsive forces. This is the so-called Coulomb explosion (see, for example, ref.[5] for experiments and [6] for its theoretical description).

When a molecular beam enters in a crystal under channeling conditions, these two effects may compete. From one hand, the vicinage effect leads to a non-additive stopping power. On the other hand, the Coulomb explosion of the molecule tends to enlarge the transversal energy of the components (the so called transverse Coulomb heating [7,8]), increasing consequently the total stopping power. The transverse Coulomb heating can occur only under channeling conditions, where it affects the ion flux distribution. In principle, both effects cannot be separated and therefore, the interplay between them in the stopping power remains still unclear. In fact, differences in stopping power results of H^+ ions and H_2^+ and H_3^+ molecules have been attributed alternatively to vicinage effects [4] or to the Coulomb explosion of the molecules [9].

In order to study the role of the Coulomb explosion in the process, we have undertaken the present experiment where, by measuring the electronic stopping power of H^+, H_2^+ and H_3^+ under channeling conditions as a function of the tilting angle, we were able not only to isolate the effect of the Coulomb explosion but also to estimate its contribution to the total stopping power.

2. Experimental setup and procedure

The measurements of the H^+, H_2^+ and H_3^+ stopping powers were done using a SIMOX target composed by a 200 nm Si ⟨100⟩ film on top of a 400 nm SiO_2 film, both being constructed over a Si ⟨100⟩ wafer. Beams of H^+, H_2^+ and H_3^+ of 150 keV/amu incident on the SIMOX target were backscattered in the target and collected by a surface barrier detector. The resolution of the detector plus the electronic system was better than 7 keV.

In the first place a H^+ channeling spectrum was obtained. With this aim we first located the ⟨100⟩ axis. Then, we tilted the sample at 6° and subsequently a complete scanning on the z axis was performed in order to identify the {100} and {110} planes. Following, a channeling spectrum was acquired at 15° with respect to the {100} plane. In the sequence, a random spectrum was recorded. This procedure was repeated at 30, 60 and 75° with respect to the {100} plane. Typical backscattering spectra are shown in figure 1 for H^+ projectiles. Finally, the whole procedure was repeated with the H_2^+ and H_3^+ molecules taking care that the energy per amu and the current per particle were the same in all experiments.

According to the procedure outlined in ref. [10], the electronic stopping power can be determined directly from the spectrum of backscattered particles by fitting a particular function that takes into account the contribution of the dechanneling particles. The results of the H^+, H_2^+ and H_3^+ channeling stopping powers as a function of the incident angle are displayed in figure 2. Each datapoint was obtained by the following procedure: in the first place, we have done the average between the four individual azimuth measurements. Then, we have taken the mean value between each pair of symmetrical tilt angles (e.g. $+0.2°$ and $-0.2°$). This mean value is the one plotted at each side of the curve shown in Fig. 2. The error bars represent the statistical uncertainty involved in the overall procedure described previously. As can be observed, the H^+ scanning shows the most pronounced dip and the most distant shoulders with respect to the center of the channel, followed by the one corresponding to H_2^+ and, finally, the shallower one belonging to the H_3^+ molecule, having the nearest shoulders.

3. Data analysis and results

For the analysis of the H_2^+ and H_3^+ stopping powers we assume that the molecular stopping power S_m as a function of the tilt angle Ψ and averaged over the azimuthal angles has three components,

$$S_m \left(H_n^+ \Psi\right) = S\left(H^+, \Psi\right) + \Delta S_{vic}\left(H_n^+\right) + \Delta S_{exp}\left(H_n^+, \Psi\right) \quad (1)$$

with $S\left(H^+, \psi\right)$ corresponding to uncorrelated H^+ fragments, the contributions due to the vicinage and the Coulomb explosion effects ΔS_{vic} and ΔS_{exp}, respectively. The sum of these last two contributions is shown in figure 3. For this sake, we have subtracted from the H_2^+ and H_3^+ distributions the one corresponding to the $S\left(H^+, \psi\right)$. Here we have used a fitting curve for $S\left(H^+, \psi\right)$ (displayed in fig. 2 with a full line) .

The contributions of the vicinage and Coulomb explosion effects on the molecular stopping power shown in figure 3 have a peculiar shape as a function of the tilt angle and can be easily interpreted by invoking the angular compensation rule proposed by Lindhard [11]. The angular average over all tilt and azimuthal angles washes out any crystalline effects and its mean value shall correspond to the one for an amorphous target. In that way, if there is an enhancement of the energy loss for $\Psi \approx 0$ due to vicinage and/or the transversal heating during the molecular breakup, there should be a compensating effect that shall lead to a decreasing of the energy loss at some other tilt angle Ψ. Physically, this decreasing at larger tilt angles can be explained in terms of a rechanneling of one of the H^+ fragments. In the case of H_2^+ projectiles at 150 keV/amu, the aperture angle (after the explosion) is about the angle where the shoulders appear.

The angular shape in figure 3 can modeled by considering the following conditions:
a) ΔS_{vic} should be nearly independent of Ψ. It has been shown in reference [4] that the vicinage effect has a weak dependence on the projectile orientation. Therefore, it basically affects the whole $S_m\,(\Psi)$ angular distribution without modifying its shape;
b) the mean value of ΔS_{exp} has to be equal to zero in accordance to the Lindhard's angular compensation rule [11] ($\int d\Psi \sin(\Psi) \Delta S_{exp}(\Psi) = 0$);
c) for larger Ψ, the function ΔS_{exp} should vanish. In non-aligned conditions, the ion flux distribution is nearly uniform. Therefore, the effect of the Coulomb explosion should be negligible;
d) the function $\Delta S_{exp}\,(H_n^+, \psi)$ at small tilt angles (smaller than the critical angle [12]) should be larger than at 0°;
e) $\Delta S_{exp}\,(H_n^+, \psi)$ should be an even function.

We found that the simplest function that fulfilled all these conditions is

$$\Delta S_{exp}\left(H_n^+, \Psi\right) = C \sin\left(k\,|\Psi| + \gamma\right) e^{-\frac{k|\Psi|(1-\cos\gamma)}{\sin\gamma}} \quad (2)$$

where C, k and γ are constants. These constants and ΔS_{vic} (tilt independent) have been used as fitting parameters.

In figure 3, with full and dashed lines the fitting of equation (2) to the H_2^+ and H_3^+ results are shown. As can be observed our proposed ansatz Eq.(2) describes very well the experimental data. The contribution of the Coulomb explosion for $\Psi = 0$ are following ones: for H_2^+, 0.18 ± 0.05 eV/Å; and for H_3^+, 0.49 ± 0.05 eV/Å. These values correspond to about 2 and 5 % of the stopping of H^+ in Si

The maximum contribution of ΔS_{exp} to the stopping power occurs at about the critical angle Ψ_c. It reaches 0.3 eV/Å for H_2 and 0.6 eV/Å for H_3. Finally, as already mentioned, the negative values of ΔS_{exp} indicate a rechanneling of one of the individual ions. The Coulomb explosion, acting as a astigmatic lens for the ion beam [6], can be the responsible for the rechanneling effect. In fact, it can redirect part of the molecule fragments that otherwise would not be channeled if the beam had no angular dispersion.

In summary, by performing a simple RBS/ channeling experiment with H^+, H_2^+ and H_3^+ projectiles on a SIMOX target we were able to evaluate the Coulomb explosion contribution to the total stopping power. With this aim, we have performed, for each angle, each azimuth, and each projectile, stopping power measurements. The experiments were repeated at four different azimuths and an average has given the H^+, H_2^+ and H_3^+ final stopping powers as a function of the incident angle. Subsequently, we have subtracted from the H_2^+ and H_3^+ stopping powers the H^+ contribution. In a next step, we have fitted the obtained results with an expression that contains the vicinage and the Coulomb explosion effects. By assuming that the vicinage effect does not affect significantly the shape of the resulting distributions and the angular compensation rule of Lindhard, we were able to extract the H_2^+ and H_3^+ Coulomb explosion contributions to the total stopping power.

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Figure Captions

Figure 1. RBS/channeling spectra of the SIMOX target. Open triangles stand for random spectrum and the squares stand for the $\langle 100 \rangle$ channeling spectrum.

Figure 2. Channeling stopping power as a function of the tilt angle. Full circles correspond to H^+ ions, open squares correspond to H_2^+ molecule and full triangles to H_3^+ molecules. The full line is the fitting to the H^+ data (using two gaussian functions) and the other lines are plotted only to guide the eyes.

Figure 3. Molecular H_2^+ and H_3^+ stopping powers after subtracting the H^+ contribution.