Energy loss measurements of H$_2$ and H$_3$ molecular beams along random and $\langle 110 \rangle$ directions of Si

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Abstract

The energy loss of H$_2$ and H$_3$ molecules along the $\langle 110 \rangle$ and non-aligned directions of Si was measured through the Rutherford backscattering technique in combination with a SIMOX target. The experiments were performed at 300, 500 and 700 keV/atom. The results show the following features: First, the random and channeling molecular stopping powers are larger than the corresponding atomic one. Second, at a given energy per atom, the H$_3$ energy loss is larger than the one of H$_2$. Third, within the experimental errors, the channeling and random molecular stopping powers are equal. Finally, there are strong indications that the contribution of the screened Coulomb explosion to the energy straggling of molecular beam increases with the molecular velocity. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

At energies where the electronic stopping power prevails, the energy loss per ion of molecular projectiles in solids can be significantly different from that of independent ions at the same velocity [1–7]. This difference has been ascribed to the so-called vicinage effect. The vicinage effect results from the simultaneous interaction of two or more ions located at short internuclear distances, moving in a correlated way through the matter. Enhancement effects on the energy loss have already been observed, but the magnitude of the effect differs significantly according to the different experiments [1–5]. In a recent work [8], by performing precise nuclear reaction analysis profiling on a set of Si$^{18}$O$_2$ films of different thicknesses, it has been shown that the ratio between the H$_3$ stopping power and the one of three uncorrelated protons reaches a maximum value of 1.8 at a thickness of 3.0 nm.

In the present work, we report measurements of the energy loss of H$_2$ and H$_3$ beams along the $\langle 110 \rangle$ axis of Si, as well as for a random direction. The measurements were accomplished by using the backscattering geometry combined with a SIMOX type of target. In this way, we were able not only...
to compare the channeling and random molecular energy losses, but also to estimate the distance of coherence and the magnitude of the contribution of the screened Coulomb explosion to the molecular energy straggling.

2. Experimental procedure and data analysis

The measurements were performed using the RBS technique along with a SIMOX target, as described in our previous work [9]. In summary, the SIMOX sample consisted of a 100 nm $\langle 100 \rangle$ Si single crystal on top of a 400 nm SiO$_2$ layer built in a Si$\langle 100 \rangle$ wafer. The roughness of the upper Si/SiO$_2$ layer was less than 2 nm, as determined by transmission electron microscopy (TEM) measurements. The sample was mounted on a three-axis goniometer with 0.01° precision, and the chamber was evacuated, reaching a final vacuum of a few $10^{-8}$ mbar.

Molecular and atomic beams at energies of 300, 500 and 700 keV/atom were produced by the 2 MV accelerator at the Groupe de Physique des Solides, Université Paris VI et VII, Jussieu, Paris. The beam divergence was less than 0.01°, and the backscattered particles were detected by a Si surface barrier detector placed at 165° with respect to the beam direction. The overall detector and electronic resolution of the system was about 7 keV (FWHM).

Since the terminal voltage of the accelerator had to be set according to the type of beam used, a shift of the beam energy per atom could occur, and, consequently, affect the results. To prevent this effect, we recorded a random RBS spectrum, whenever the type of beam was switched. Through the position of the Si front edge, we were capable of reproducing the required energy per atom with an uncertainty of about 125 eV/atom. In any case, small shifts in the position of the Si front edge were taken into account for the data analysis.

The difference $\Delta E_n$ in the position of the Si/SiO$_2$ RBS edge, as measured through molecular (H$_n$ and H$_3$) and protons beams, gives the difference in the energy lost along the inward path. On the other hand, from the comparison between the widths of the Si/SiO$_2$ edges when measured by molecular and atomic beams, one can obtain the contribution of the screened Coulomb explosion $\sigma_{\text{Coul}}$ to the energy straggling of the molecular beam.

3. Results and discussion

In Fig. 1, we display 500 keV/atom random RBS spectra of a SIMOX sample measured with protons and H$_3$ beams. As can be observed, the Si/SiO$_2$ edge of the spectrum taken with H$_3$ is shifted towards lower energies with respect to the one determined by protons. This feature already shows that the random molecular stopping power per atom is larger than the atomic one. On the other hand, the edge corresponding to the Si/SiO$_2$ interface is wider in the spectrum obtained with H$_3$. This effect is due to the contribution of the screened Coulomb explosion of the molecule to the energy straggling. Fig. 2 presents similar spectra to the ones shown in Fig. 1, but for incidences aligned with the $\langle 110 \rangle$ channel. Again, the same kind of characteristics are present.

Table 1 presents a summary of all our results. The difference $\Delta E_n$ in the energy lost by molecular (H$_n$) and atomic ions backscattered at the Si/SiO$_2$ interface, for both random and channeling trajectories, are displayed in the third and fourth columns, respectively. Each $\Delta E_n$ value is the result of
The superscript r and c stand for random and channeling, respectively. For the
see review by Arista [6]. However, those obtained
the literature for C and Au self-supported films –
are in agreement with those previously reported in
ditions. The results obtained for random direction
mentioned above, the absence of a systematic and
random direction. However what is surprising is, as
Summary of the results obtained in the present work
Table 1

<table>
<thead>
<tr>
<th>Energy (keV/atom)</th>
<th>Ion</th>
<th>ΔE⁺ (keV)</th>
<th>ΔE⁻ (keV)</th>
<th>L⁺ (nm)</th>
<th>L⁻ (nm)</th>
<th>λ (nm)</th>
<th>σ⁺\text{Coul} (keV)</th>
<th>σ⁻\text{Coul} (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>H⁺</td>
<td>0.6 ± 0.2</td>
<td>0.5 ± 0.2</td>
<td>17 ± 6</td>
<td>14 ± 6</td>
<td>40</td>
<td>1.5 ± 0.4</td>
<td>1.2 ± 0.4</td>
</tr>
<tr>
<td></td>
<td>H₂⁺</td>
<td>1.3 ± 0.4</td>
<td>1.6 ± 0.4</td>
<td>18 ± 6</td>
<td>22 ± 6</td>
<td></td>
<td>2.4 ± 0.1</td>
<td>2.5 ± 0.2</td>
</tr>
<tr>
<td>500</td>
<td>H⁺</td>
<td>1.1 ± 0.3</td>
<td>0.8 ± 0.3</td>
<td>41 ± 11</td>
<td>30 ± 11</td>
<td>66</td>
<td>2.3 ± 0.1</td>
<td>1.8 ± 0.2</td>
</tr>
<tr>
<td></td>
<td>H₂⁺</td>
<td>1.9 ± 0.5</td>
<td>2.0 ± 0.5</td>
<td>36 ± 10</td>
<td>38 ± 10</td>
<td></td>
<td>3.6 ± 0.5</td>
<td>3.5 ± 0.4</td>
</tr>
<tr>
<td>700</td>
<td>H⁺</td>
<td>0.3 ± 0.1</td>
<td>0.3 ± 0.1</td>
<td>14 ± 5</td>
<td>14 ± 5</td>
<td>92</td>
<td>2.4 ± 0.7</td>
<td>2.3 ± 0.7</td>
</tr>
<tr>
<td></td>
<td>H₂⁺</td>
<td>1.2 ± 0.2</td>
<td>0.9 ± 0.2</td>
<td>27 ± 5</td>
<td>20 ± 5</td>
<td></td>
<td>3.6 ± 0.4</td>
<td>3.8 ± 0.4</td>
</tr>
</tbody>
</table>

The superscript r and c stand for random and channeling, respectively.

As molecules penetrate into a target, the distance between the atomic components increases as a consequence of both the screened Coulomb explosion and the multiple scattering. The thickness of the Si upper layer of our SIMOX sample is such that, due to these two processes, the molecular contribution to the stopping power becomes negligible much before the components reach the Si/SiO₂ interface. Thus, the ΔEₙ values that we have measured reflect both the variation of the molecular effect with the distance between atomic components and the variation of this distance with penetration depth. We have chosen to use the experimental ΔEₙ values to evaluate a lower limit for the coherence distance, that is, the distance along which the H atoms in the molecule act in a coherent way. For this purpose, we assume that all over the distance L, the contribution of the molecular effect to the energy loss is constant, and equal to its value at the target entrance. Within this assumption, this distance can be obtained from ΔEₙ through the following expression:

\[
ΔEₙ = K(S(Hₙ)/n - S(H))L,
\]

where K is the kinematic factor for backscattering of proton by silicon, and S the stopping power. The proton energy loss was taken from the TRIM subroutine RSTOP [10], which provides random stopping power. This procedure was followed even for the channeling case, as the distance of coherence is not large enough for the beam to attain statistical equilibrium. Regarding the molecular stopping power, we used the maximum energy loss enhancement predicted by the equilibration rule [11],

![Graph showing yield vs channel for 500 keV/atom]
namely: $S(H_3)/3S(H) = 2$ and $S(H_2)/2S(H) = 1.5$ [1], so that the Eq. (1) yields an underestimation for the distance of coherence. The results are shown in the fifth and sixth columns of Table 1. They are in agreement with previous measurements carried out through the resonant $^{18}$O(p,α)$^{15}$N nuclear reaction at 151.2 keV [8].

The experimental $L$ values that we have derived can be compared to a simple estimate based on the two following assumptions. The first one is that the vicinage effect vanishes for internuclear distances larger than the adiabatic radius $R_{ad} = \frac{\lambda}{\omega}$, where $v$ is the ion velocity, and $\omega$ the plasma frequency. The second assumption is that the separation between atomic components up to $R_{ad}$ is determined solely by the Coulomb repulsion, and thereby we neglect multiple scattering. Since multiple scattering is expected to be less important under channeling, this assumption is supported by the fact that there is no significant difference between the $\Delta E_n$ values for random and channeling trajectories. This estimate should lead to an upper limit for the penetration depth over which molecular effects are observed. We shall call $\lambda$ this upper limit.

According the second assumption, the separation between the ions grows form the initial distance $R(0) = R_0$ to the distance $R$ at the time [1]

$$t = t_0 \left\{ \xi^{1/2}(\xi - 1)^{1/2} + \ln \left[ \xi^{1/2} + (\xi - 1)^{1/2} \right] \right\},$$  

(2)

where $t_0 \equiv (\mu R_0^2/2Z_i^2e^2)^{1/2}$, $\mu$ is the reduced mass of the molecule, and $\xi = R/R_0$. Thus, the maximum distance of coherent $\lambda$ can be determined, if in $\lambda = vt$, $t$ is taken from Eq. (2) with $R = R_{ad}$. Consequently, we expect that the experimental coherence distance, which – as explained above – are necessarily an underestimation, should be somewhat smaller than $\lambda$. The values for $\lambda$ calculated according to Eq. (2) are displayed in the seventh column of Table 1. It is worth mentioning that this calculation is valid only for $H_2$. A comparison with the experimental distances $L$ shows that, for all the cases, they are smaller than $\lambda$.

The contribution of the Coulomb explosion to the straggling of the molecular beams can be observed in Table 1 (columns 8 and 9) as well. An inspection of those results show that within the experimental errors this contribution increases with the velocity of the molecule. Again the difference between random and channeling conditions are within the experimental errors and are not systematic.

4. Conclusions

We performed measurements of the energy loss of $H_2$ and $H_3$ molecules along the Si(110) channel as well as for a non-aligned directions. To this end, we used the Rutherford backscattering technique in a SIMOX target. The results show that: (a) channeling molecular energy losses are larger than the corresponding atomic one; (b) at a given energy per atom, the $H_3$ energy loss is larger than the one of $H_2$; (c) there is no significant difference between the channeling and random molecular energy losses.

A lower limit for the distance along which the ions in the molecule interact coherently with the target atoms – the distance of coherence – was calculated from our experimental data. In the energy range investigated, this distance lies between 15 and 40 nm.

Finally, it has been found that, within the experimental errors, the contribution of the screened Coulomb explosion to the energy straggling seams to be proportional to the molecule velocity, and is about the same for random and aligned incidences.

Acknowledgements

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References