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Energy loss measurements of H_2 and H_3 molecular beams along random and $\langle 1\,1\,0\rangle$ directions of Si

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Abstract

The energy loss of H_2 and H_3 molecules along the $\langle 1\,1\,0\rangle$ 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.

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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 $\langle 1\,1\,0\rangle$ 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

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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 ΔE_n in the position of the Si/SiO_2 RBS edge, as measured through molecular (H_2 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 σ_{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 ΔE_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 ΔE_n value is the result of

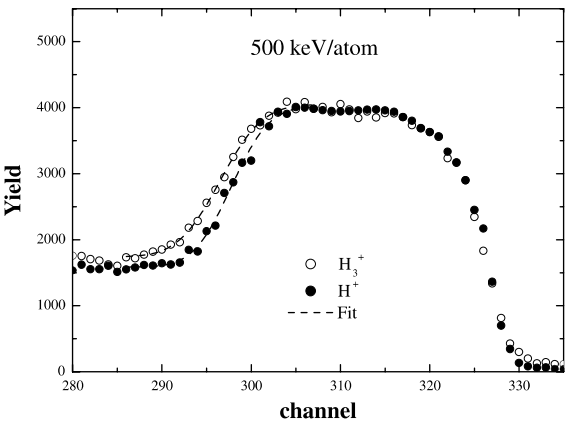

Fig. 1. RBS random spectra of a SIMOX target measured with 500 keV/atom H and H_3 beams. The dashed lines are the results of fitting to the Si/SiO_2 interface region.

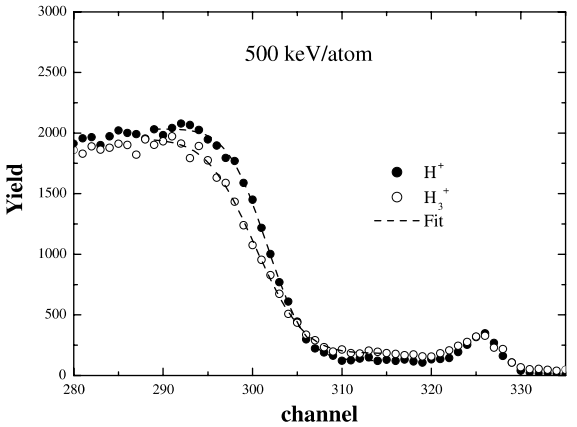

Fig. 2. The same as Fig. 1, but for an incidence aligned with the $\langle 1\,1\,0\rangle$ axis.

an average over three independent measurements. From the values obtained, one can see that, for random as well for channeling conditions: (a) the molecular (H_2 and H_3) energy loss per atom are larger than the corresponding atomic one; (b) at the same energy per atom, the H_3 energy loss per atom is larger than the H_2 one; (c) the maximum ΔE_n value was obtained at 500 keV/atom; and (d) there is neither significant nor systematic difference in ΔE_n between the random and the channeling conditions. The results obtained for random direction are in agreement with those previously reported in the literature for C and Au self-supported films – see review by Arista [6]. However, those obtained for the $\langle 1\,1\,0\rangle$ direction are original ones. They follow the same tendency as the ones obtained in random direction. However what is surprising is, as mentioned above, the absence of a systematic and significant difference between the results obtained in both kind of measurements.

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/SiO_2 interface. Thus, the ΔE_n 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 ΔE_n 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 ΔE_n through the following expression:

$$\Delta E_n = K(S(\mathrm{H}_n)/n - S(\mathrm{H}))L, \tag{1}$$

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],

Table 1
Summary of the results obtained in the present work

Energy (keV/atom)	Ion	ΔE^{r} (keV)	ΔE^{c} (keV)	L^{r} (nm)	L^{c} (nm)	λ (nm)	$\sigma^{\mathrm{r}}_{\mathrm{Coul}}$ (keV)	$\sigma^{\mathrm{c}}_{\mathrm{Coul}}$ (keV)
300	H_2^+	0.6 ± 0.2	0.5 ± 0.2	17 ± 6	14 ± 6	40	1.5 ± 0.4	1.2 ± 0.4
	H_3^+	1.3 ± 0.4	1.6 ± 0.4	18 ± 6	22 ± 6		2.4 ± 0.1	2.5 ± 0.2
500	H_2^+	1.1 ± 0.3	0.8 ± 0.3	41 ± 11	30 ± 11	66	2.3 ± 0.1	1.8 ± 0.2
	H_3^+	1.9 ± 0.5	2.0 ± 0.5	36 ± 10	38 ± 10		3.6 ± 0.5	3.5 ± 0.4
700	H_2^+	0.3 ± 0.1	0.3 ± 0.1	14 ± 5	14 ± 5	92	2.4 ± 0.7	2.3 ± 0.7
	H_3^+	1.2 ± 0.2	0.9 ± 0.2	27 ± 5	20 ± 5		3.6 ± 0.4	3.8 ± 0.4

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

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 $^{18}O(p,\alpha)^{15}N$ 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 $R_{ad} = \frac{v}{\omega_0}$, where v is the ion velocity, and ω_0 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 ΔE_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 λ this upper limit.

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

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

where $t_0 \equiv \left(\mu R_0^3/2Z_1^2e^2\right)^{1/2}$, μ is the reduced mass of the molecule, and $\xi = R/R_0$. Thus, the maximum distance of coherent λ 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 λ. The values for λ 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 λ.

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⟨1 1 0⟩ 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.

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