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Coulomb Heating Behaviour of Fast Light Diclusters in Si<100> Direction

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1. Introduction

Beams of molecules and cluster ions are useful tools in fundamental research with promising applications in materials science and plasma physics. In particular, significant coherence effects (vicinage effects) have been predicted theoretically and in some cases experimental molecular stopping forces show clear deviations from simple additivity rules concerning the projectile constituents (Arista, 2000; Brandt et al., 1974). Other effects related to the correlated motion of cluster atoms, such as Coulomb explosion (Gemmell et al., 1975), enhanced electron emission (Rothard et al., 1990), desorption and sputtering (Matsuo et al., 1997; Yamagushi et al., 1995) have also been reported and reviewed in recent publications (Yamada et al., 2003).

In the case of crystalline materials, ions entering nearly parallel to a particular crystal axis or plane become channelled as their motion is guided by correlated collisions with target atoms. The average transversal momentum of channelled atomic particles increases due to the inelastic scattering with the target electrons and scattering at displaced target atoms. This effect enhances the number of close encounters with the atomic rows and is named transverse heating. It has been observed that charge-changing processes of fast heavy ions may even lead to transversal cooling, namely a reduction of the transversal energy (Assmann et al., 1999).

In addition to the channelling motion, molecular ions undergo a break-up process, since they lose their bonding electrons due to ionisation in the first monolayers of the material. The combination of these two correlated motions, namely the ion channelling and the break-up of the cluster under quasi-Coulomb forces leads to a transverse *Coulomb heating*. Pioneering investigations of this Coulomb heating have been performed using the transmission technique. (Poizat et al., 1972). On the other hand by measuring the energy spectra of backscattered protons from H⁺, H₂⁺, and H₃⁺ beams (Caywood et al., 1971; Tombrello et al., 1973) they were able to deduce the dynamics of the molecular break-up process during channelling effects in Si. But no attempt was done to quantify the Coulomb heating. Only much later the Coulomb heating was at last estimated from dechannelling profiles of hydrogen molecules. However, the computer simulations employed in such study strongly overestimated the experimental results. (Khodyrev et al., 2002).

In this work we review on a quantitative determination of the transverse Coulomb heating energy (abbreviated *Coulomb heating*) for H and H₂, B and B₂, and C and C₂ beams irradiated on the Si<100> direction. To this end, we used the Si-K α x-ray production and also the backscattering yield (RBS-C) of the mentioned single and molecular beams channelling along the aforementioned direction. Since vicinage effects do not affect the K-shell x-ray and backscattering yields, because the characteristic impact parameters for both processes are much smaller than the typical distance for vicinage effects, differences between experimental results for single and molecular beams are directly linked to the Coulomb explosion during the channelling motion. The experimental results for the Coulomb heating were compared to results of advanced computer simulations being the agreement quite good. These simulations were done in order to understand the physics lying behind the Coulomb heating effects.

In the present chapter we will show the experimental results obtained for H and H₂, B and B₂, and C and C₂ beams. In the case of the last two beams the energy /amu was changed in a wide interval and the obtained Coulomb heating results as a function of the potential energy stored in each ion of the molecule seems to follow on a straight line indicating some kind of “universal behaviour”.

2. Experimental procedures

The experiments were carried out at the Ion-Implantation Laboratory of the Federal University of Rio Grande do Sul, Porto Alegre, Brazil. In all cases, monoatomic and cluster beams were irradiated on crystalline silicon targets after the alignment procedure and both backscattered ions and x-ray emissions as a function of the tilt angle were simultaneously detected and recorded.

In what follows, the experimental procedures are described according to the ion/cluster combination used.

2.1 H and H₂ beams

The experiments were carried out at the Ion-Implantation Laboratory of the Federal University of Rio Grande do Sul. The 500 kV electrostatic accelerator has delivered beams of H⁺ and H₂⁺, at 150 keV/amu with an average current reduced down to about 100 nA for H⁺ and 50 nA for H₂⁺ (these currents are typically smaller than the ones used for ion implantation). In order to ensure identical conditions, we kept the same proton flux hitting the target for all particles and clusters under study. Furthermore, the currents were reduced by an additional factor of 10 in the case of backscattering measurements. The Si (100) crystal was mounted on a goniometer for the alignment procedure (Azevedo et al., 2002). We have fixed the azimuthal angle $\Phi = 22.5^\circ$ (relative to the plane [100]) in order to avoid a full azimuthal averaging procedure around the <100> channel, which would be time consuming and could lead to unacceptable damage of the sample.

The backscattered particles were detected by a surface-barrier detector located at 165° with respect to the incident beam. The overall resolution of the detector plus the electronic system was for protons better than 7 keV. The 1.74 keV Si K α x-rays emitted from the target were detected by Si(Li) detector with an energy resolution of 180 eV at 5.9 keV. This detector was mounted at 135° with respect to the incident beam. This constrained the measurements

only to directions around the Si <100> one. Once the center of the channel was determined, a sweep from -5° to $+5^\circ$ was performed in steps of 0.2° around the center of the channel. At each stage the corresponding RBS of the incident beam and the yield of the x-ray produced by the Si target were recorded.

The vacuum system of the analysing chamber consists of an oil-free turbomolecular drag pump with a liquid nitrogen trap, capable of reaching a final pressure below 10^{-7} mbar. It is important to point out that the samples were cleaned and etched using 10% HF before each measurement to remove the native surface oxide, in order to ensure a better channelling procedure.

2.2 C, C₂ and B, B₂ beams

In these cases the incident beams were provided by the 3 MV Tandatron of the Ion-Implantation Laboratory of the Universidade Federal do Rio Grande do Sul. The experimental procedure was basically the one that was described in section 2.1 with two major differences: first, the samples were mounted on a four axis goniometer and the channelling procedure was performed with He beams of 1.2 MeV following the same procedure as described above; second, subsequently the position of the sample was changed by using the vertical axis of the goniometer in order to get a fresh point not previously damaged by the alpha beam. Following we have changed to a C⁺ (or B⁺) beam and performed the sweep around the channel <100> and recording at each 0.2° the corresponding RBS and x-ray yield following the procedure described above. Once this procedure was finished we have changed to a C₂⁺ (or B₂⁺) beam. On this occasion we moved again the sample to another fresh point of the sample free of damage and we have repeated the above-described procedure.

As can be observed from the mentioned description an extreme care has been taken in order to avoid or minimize the damage induce by the incoming beam, during the scanning procedure. These procedures have to be taken because the x-ray emission is very sensitive to the damage or amorphization produced in the channel.

In the present case the overall resolution of the detector and electronic system was around 30 keV for C and 25 keV for B ions (FWHM) respectively.

It should be mentioned that for C, C₂ and B, B₂ the energy interval spanned in the present experiment went from 900 keV/atom up to 2200 keV/atom, for B and from 800 to 2200 keV/atom for C respectively.

For each beam energy, the x-ray yields as well the RBS-C spectra were obtained as an average from five up to six (depending on the RBS-C cross section) independent measurements performed under the same condition of ion flux. The uncertainties were calculated taking into account the independent measurements as well as the fitting to the spectra (to be described in Section 3).

3. Experimental results

3.1 H and H₂ data treatment and results

Measurements of close encounter events giving raise to x-ray emission and backscattering under channelling conditions provide a sensitive method to investigate

variations and redistribution of the ion flux close to a row of atoms and can be used to quantify the heating process inside crystal channels. The results for the x-ray emission induced by H and H₂ beams at 150 keV /atom are depicted in Fig. 1 (Fadanelli et al., 2004) as a function of the projectile entrance angle. Here, large tilt angles correspond to nearly random direction. As can be observed from the figure, atomic and molecular beams at the same energy, flux and fluence per atom act differently in the Si induced x-ray production.

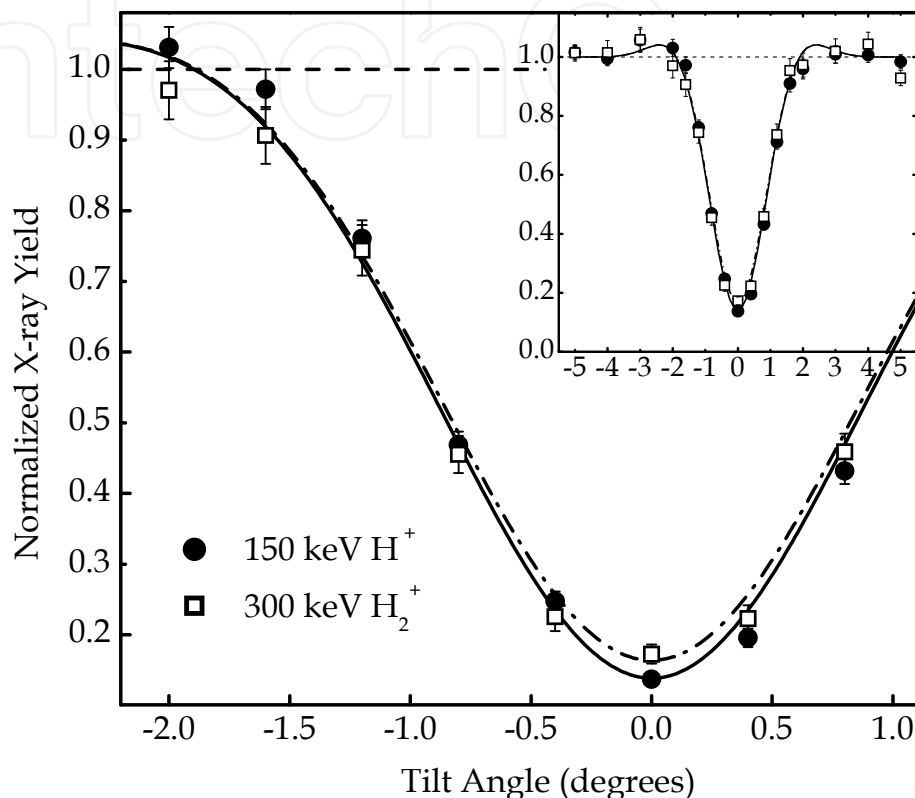


Fig. 1. Si x-ray emission as a function of the tilt angle induced by H and H₂ beams. In the inset is shown the results of the whole sweep obtained between -5° and +5° (Fadanelli et al., 2004).

First it should be emphasized that we do not observe any molecular effects at random direction (within 2% of difference), feature that indicates that no interference molecular effects, like the vicinage one, affect the results of the present experiments. On the other hand there are significant differences at small polar angles. In comparison to H, the yield for H₂ is 26% larger at the central channel direction.

In order to obtain quantitative results from the experiments for the Coulomb heating, it is necessary to find the dechanelling rate as a function of the tilt angle $\alpha(\psi)$. Since the Coulomb explosion time is about one order of magnitude smaller than the oscillation period time of the ion inside the channel, the Coulomb heating can act as an “astigmatic lens” for the channelled ions (Sigmund, 1992). In order to extract the experimental values for the Coulomb heating we have used the two-beam model (Götz et al., 1988; Lindhard, 1965), which have been successfully used in previous works (dos Santos et al., 1997; Fadanelli et al., 2004). The two-beam model is, in fact, a particular case of the discontinuous model used to solve the master equation for the transverse-energy distribution (Götz et al., 1988). In this

model, the beam is considered as a sum of two beam fractions distinguished by their transverse energy range: the fraction of ions below a critical value is called “channelled”. The remained fraction is called “dechannelled”. Under the restriction of a nearly perfect crystal we have that the dechannelled fraction $\chi(z)$ is given by:

$$\frac{d\chi}{dz} = \alpha[1 - \chi(z)] \quad (1)$$

where z is the depth inside the channel and $[1 - \chi(z)]$ is the channelled fraction which as well as $\chi(z)$ are both function of the penetration depth z . The parameter α is a weak function of the energy, so in agreement with previous works we have taking as independent of the energy in the studied depth range of 400 nm. We have calculated that in this zero approximation the final error in the Coulomb heating results is less than 5%. The α value was determinate for each incident angle ψ from the fitting of the RBS-C spectra. For this analysis we have used the electronic stopping to convert the energy loss in depth z as well as the Rutherford cross section to determine the backscattering yield (Chu et al., 1978) and each spectrum was convoluted with the experimental resolution and energy loss straggling. To describe the surface peak, we have considered a thin random layer in the surface.

In order to determine the H_2 corresponding α parameters the RBS-C fittings were obtained following the same procedure as described above. In Figure 2 are shown two well

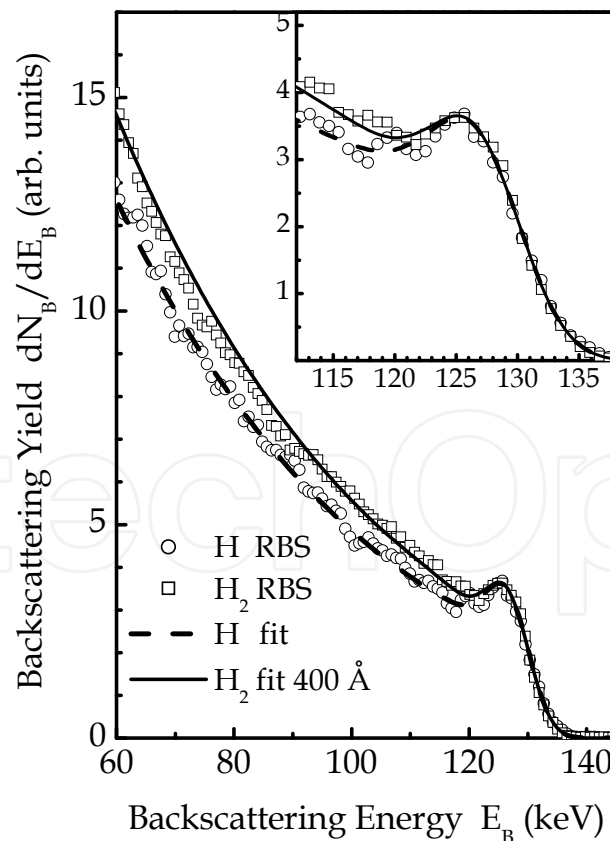


Fig. 2. RBS yield as a function of the energy. Curves stand for the fitting procedure according to the two-beam model assuming that the Coulomb explosion effectively takes place at a critical depth of 400 Å (Fadanelli et al., 2004).

channelled H and H₂ RBS-C spectra (Fadanelli et al., 2004). It can be clearly seen that the molecular RBS-C spectrum shows an increasingly larger dechannelling (for lower backscattering energies) than the corresponding to the H one. In this kind of experiments the dechannelling due to the Coulomb explosion seen in RBS spectra is more striking than the one observed in the x-ray experiments. This happens because as will be described below there is a delay in the Coulomb explosion effect. Consequently, the dechannelling effect is also a clear signature of the Coulomb heating effect.

At this point it should be mentioned that the Coulomb heating effect does not occur immediately after the molecule enters into the sample. In fact the molecular break-up begins soon after the molecular ions penetrate a few monolayers in the target. However the Coulomb explosion usually takes several hundreds of angstroms of ion penetration before changing the ion flux and thus the transverse energy distribution. The fitting of the RBS-C spectrum of the H₂ molecule, after few angstroms does not reproduce the experimental results. Although the flux change is a continuous process, for simplicity we will assume in the fittings that before a certain distance Δz the molecular beam flux is the same as the monoatomic one and that all changes in flux due to the explosion are complete after a certain Δz . Consequently, the obtained Δz values are a first-order estimation of the heating delay.

In this sense in order to obtain a good fit we have assumed that Δz is different from zero. However, we have obtained several possible $(\alpha(\psi), \Delta z)$ yielding a reasonable fit to the molecular spectra. Then, we fitted the x-rays integrated results as well (not shown in Fig. 1) to ensure that a correct pair is chosen. The two-beam model function was modified to take into account the K-shell ionisation cross sections. Finally a unique pair $(\alpha(\psi), \Delta z)$ that fits simultaneously the x-ray yields and the RBS-C spectra was found. In the present case the obtained Δz value was 400 angstroms.

When the $\alpha(\psi)$ is known, it could be found the Coulomb heating energy through the Hamiltonian of the transverse ion motion which, for small ψ angles, reads

$$E_T = E\psi^2 + U(x, y) \quad (2)$$

where E_T is the transverse ion energy, E is the ion initial energy and $U(x, y)$ is the channelling potential. Starting from Eq. (2) and taking into account the mentioned features of the cluster ion fragmentation, the Coulomb heating energy is finally given by

$$Y_{molecular}(\psi) = Y_{atomic} \left(\sqrt{\psi^2 + \Delta E_c/E} \right) \quad (3)$$

where $Y_{molecular}$ is the x-ray or RBS cluster ion yield, Y_{atomic} is the monoatomic ion corresponding one, and ΔE_c is the Coulomb heating energy. Therefore, in order to determine ΔE_c , it is necessary to determine $\alpha_{molecular}$ in the center of the channel and the value $\Delta\psi$ that leads to $\alpha_{atomic}(\Delta\psi) = \alpha_{molecular}(0)$, as shown in Fig. 3. Therefore, from Eq. (3), we have $\Delta\psi = \sqrt{\Delta E_c/E}$.

Then the experimental Coulomb heating of H₂⁺ at 150 keV/amu obtained from the present experiment is 2.6 ± 0.6 eV. This value is significantly smaller than the estimated free

Coulomb heating (4.5 eV), indicating either a target screening or a molecule alignment inside the target.

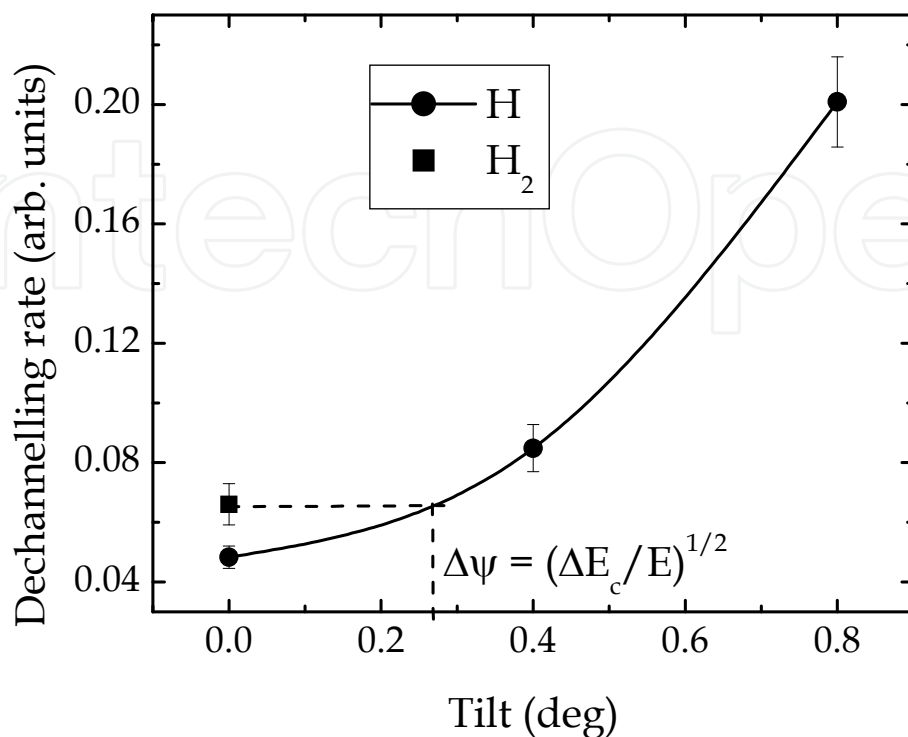


Fig. 3. Dechannelling rate as a function of the tilt angle obtained via TBM fittings of RBS spectra. The depth delay Δz used in this example is 400 Å.

3.2 Heavier ions data treatment and results

In what follows, only a brief summary of B_2^+ and C_2^+ results are shown. The data treatment is essentially identical to the one used for H_2^+ . The main difference, besides the ion type, is the different range of energies used, instead of one single ion energy as done for H_2^+ . Again, we do not observe any molecular effects at random direction (within 2% of difference) in RBS or x ray measurements.

In the aligned direction, however, it is possible to observe an increase of the x ray, as shown in Figure 4, and RBS yields, as depicted in Figure 5, (Fadanelli et al., 2008) for the diatomic ions. This is the signature of Coulomb heating for the heavier diatomic projectiles. For each dicluster it is possible to see an increase of the Coulomb heating effect as a function of energy. This behaviour indicates an important role played by the screening inside the target.

In Figure 5 it is shown both the raw experimental results for RBS yields (symbols) and the fitting procedures not taking into account the depth delay for the explosion (dashed line) and considering the delay (full line). The beam is aligned with the channel <100>. As in case of H^+ and H_2^+ , to describe the surface peak, we have considered a thin random layer in the surface (about 20 Å). The importance of the delay is emphasized by comparing the dashed and the full lines.

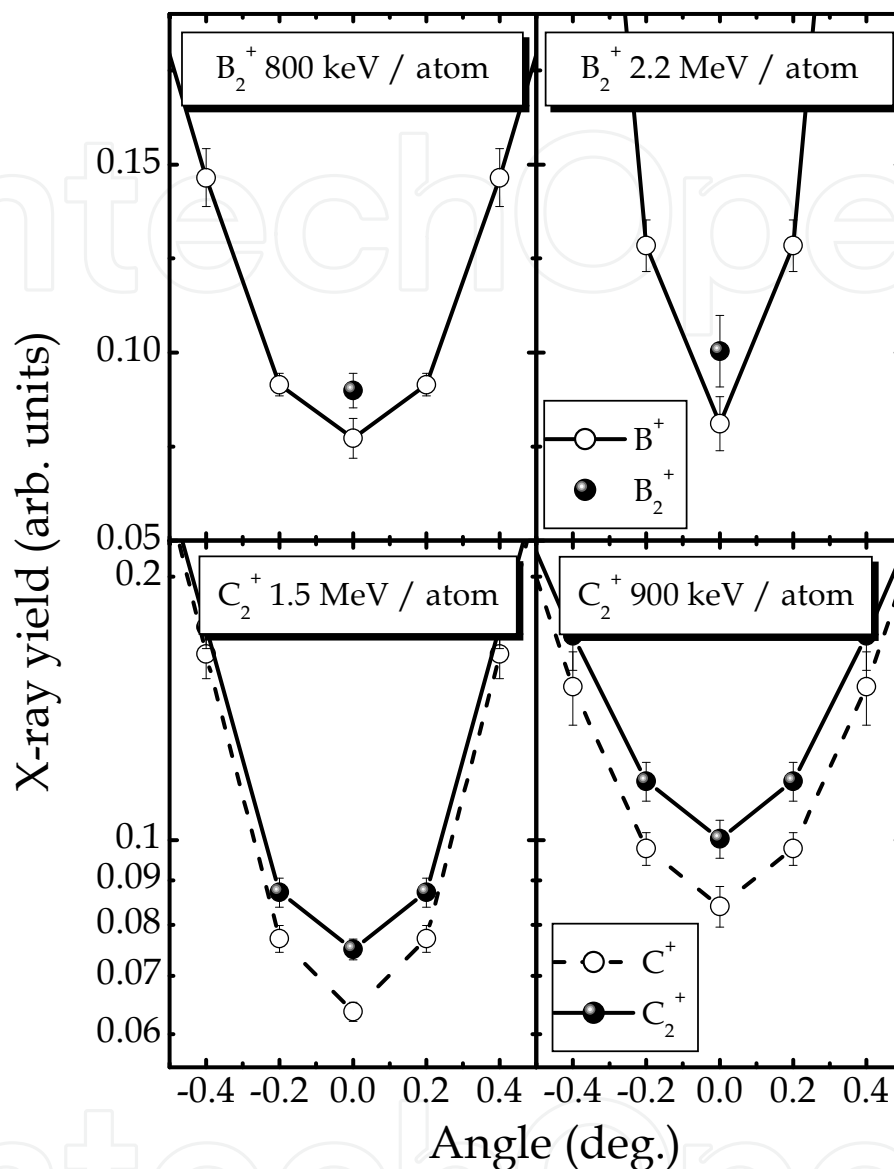


Fig. 4. Si x-ray yields near the center of the Si(100) direction. It is clear that the Coulomb heating signature for B_2^+ and C_2^+ increases with the energy and the atomic number of the molecule constituents (Fadanelli et al., 2008).

After performing essentially the same data treatment procedure as used for H_2^+ , we show in Figure 6 the Coulomb heating results for B_2^+ and C_2^+ (Fadanelli et al., 2008; Fadanelli et al., 2010). As can be observed, in both cases the Coulomb heating is a function of the corresponding beam energy. For B_2 the Coulomb heating raises from 9 eV up to 17 eV. Instead for C_2 it increases from 14 up to 30 eV. At the same time the Coulomb delay explosion is also a function of the beam energy and ranges from 1200 Å up to 2500 Å. Details about the simulation results will be given in Section 4.

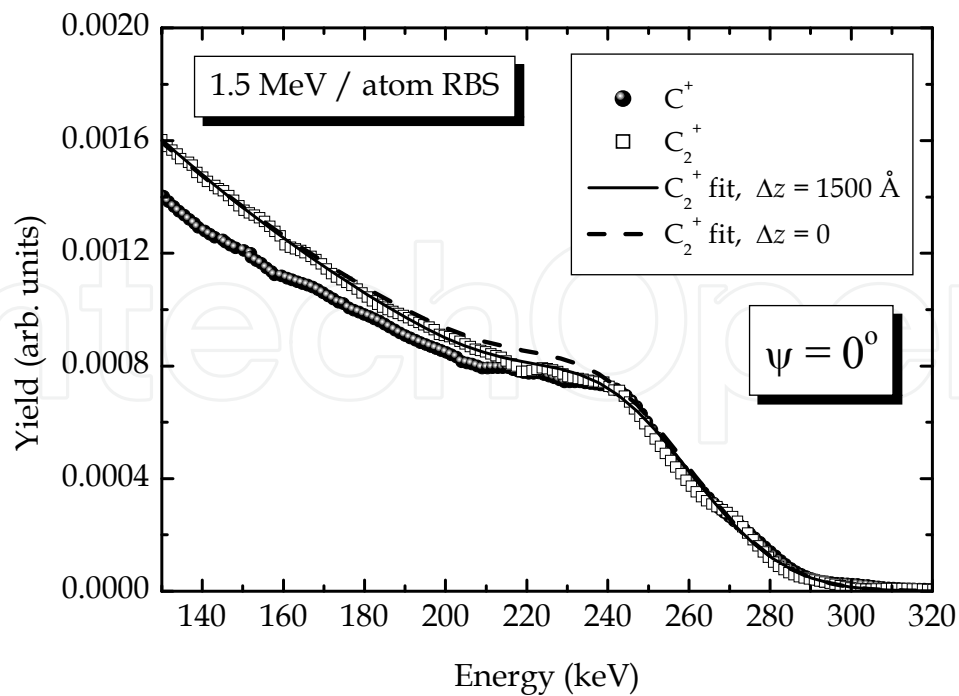


Fig. 5. C^+ and C_2^+ RBS spectra at 1500 keV / atom, taken under alignment conditions (Fadanelli et al., 2008).

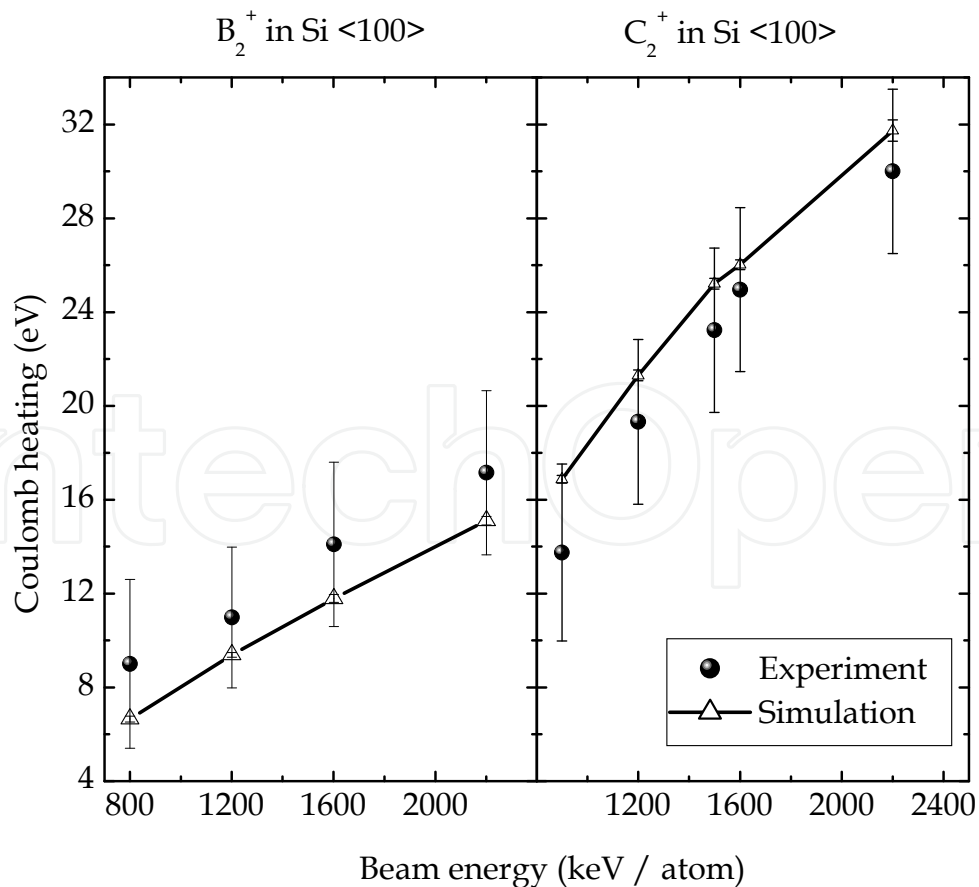


Fig. 6. Coulomb heating results for B_2^+ and C_2^+ (Fadanelli et al., 2008, 2010).

4. Simulations

Classical Monte Carlo simulations were performed for all corresponding experimental cases. To this end, we have modelled the interaction between the ions and the Si<100> target atoms using Molière continuum potential (Lindhard, 1965 & Gemmell, 1974) generated by sixteen strings, including a correction to take into account the thermal vibrations (Eckstein, 1991). The ions were allowed to move inside the channel defined by the four central strings. The interactions among the molecule fragments were modelled using either Yukawa potential (for H_2^+) or, if the number of electrons bound to the ion is large, static Dirac-Hartree-Fock-Slater potentials (Salvat et al., 1987).

In order to include a non-uniform energy loss, it was used the unitary convolution approximation (UCA) (Azevedo et al., 2000; Schiwietz et al., 1999). UCA results consist in energy loss values as a function of the distance between a given ion and the channel strings. The angle between the molecular axis and the motion direction, as well as the initial position of the projectile impinging the channel are chosen randomly. The ion charge state is chosen randomly from a gaussian distribution centred in the equilibrium charge value Z_{eq} with known width Δq (Schiwietz et al., 2001). Due to the relatively small amount of experimental information about B and C equilibrium charge states under channelling conditions in our energy range, we have selected the interpolation formulae of Ref. (Schiwietz et al., 2001), which corresponds to the ones obtained for amorphous Si. This choice seems reasonable, since in Ref. (Jiang et al., 1999a, 1999b) have investigated equilibrium charge states for different ions impinging on Si films under channelling and random conditions. In particular Jiang group found that for N at energies similar to the present ones or higher, the equilibrium charge states were achieved and are independent on the channelling or random conditions of the target film. The charge state is, finally, kept constant along all considered ion path and the Molière potential screening parameter for a given ion path is evaluated accordingly. It is important to point out that, in this way, charge changing effects along each ion motion were not taken into account in our simulations.

In this procedure we assume that some of the electrons from the impinging B_2^+ and C_2^+ molecules are suddenly removed along the first monolayers and, consequently, the molecule undergoes a break-up process. We assume that each ion reaches its own equilibrium charge state as soon as the break-up process begins. In this way, our simplified simulation does not take into account a more detailed molecular fragmentation description. In fact, orientation, velocity and position-dependent effects on the molecule break-up (as, for instance, wake effects (Arista, 2000)) not included here are expected to decrease the obtained theoretical values for the Coulomb heating.

In order to study the explosion effects on the ion flux, the simulations were carried out only along the first 4000 Å. There are two reasons for this depth choice. First, at 4000 Å, it is safe to assume that the potential energy stored in the molecule is nearly completely transferred to the molecular fragments motion. Thus, no further increase on the transverse energy can be expected from the Coulomb explosion after that depth. Second, a significant amount of ions is still channelled. Therefore, the ion flux inside the channel is still concentrated near the channel center, originating the so-called flux peaking.

Fig. 7 shows, as an example, the ion flux for both C and C_2 beams (Fadanelli et al., 2008) averaged along the depth (4000 Å). The ion flux for the C^+ beam displays the well-known

flux peaking, i.e. a strong enhancement of the flux near the center of the channel. The C_2^+ beam still shows the flux peaking effect. However, as presented in Figure 7, this effect is considerably damped for the molecular beam due to the Coulomb explosion, which enhances the transverse energy of the molecule fragments and, consequently, produces higher ion fluxes far from the channel center.

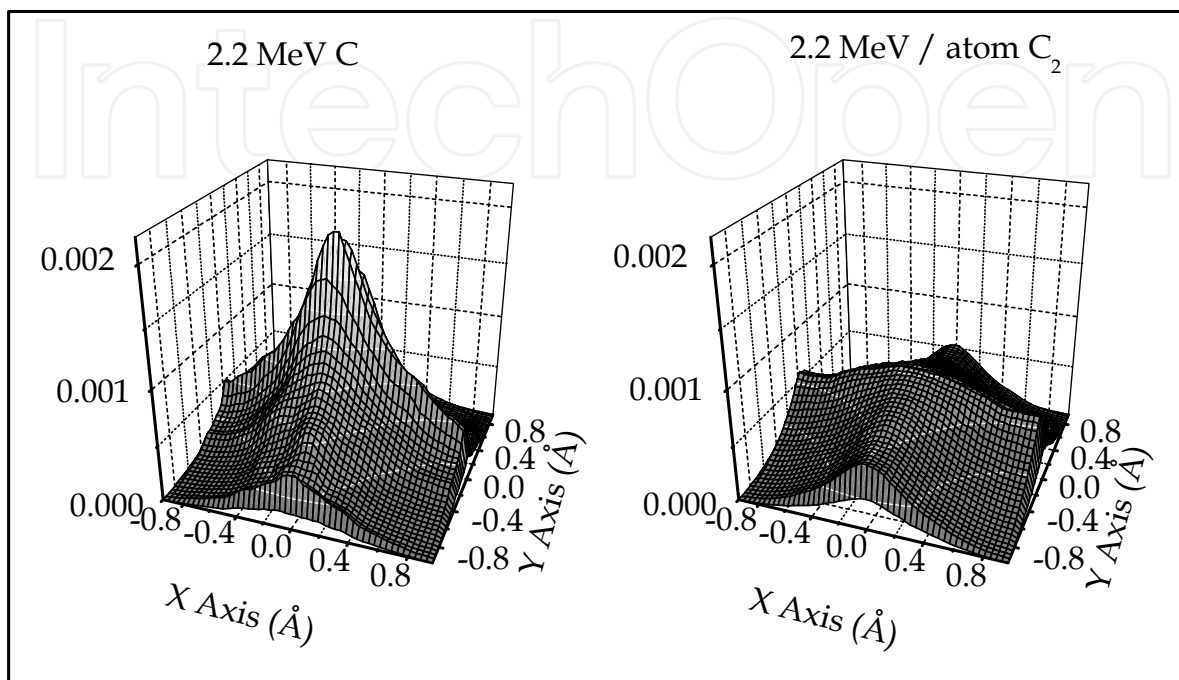


Fig. 7. C and C_2 fluxes inside the <100> Si channel obtained via simulation of aligned 2.2 MeV / atom C and C_2 beams (Fadanelli et al., 2008).

In order to evaluate the average transverse explosion energy transfer to the molecule fragments, we considered, for each beam energy, the distributions of transversal energies at 4000 Å depth in the target, far after the Coulomb explosion process. Two of these distributions, as obtained from our simulations (Fadanelli et al., 2004, 2008), are shown in figure 8 which corresponds to 150 keV / atom H^+ and H_2^+ (left panel) and 2.2 MeV/atom C^+ and C_2^+ beams (right panel). As can be observed for the same yield, the transversal energy distributions for the molecular ions are shifted to higher energies, a consequence of the Coulomb-heating process. The theoretical values of the Coulomb heating were then obtained by shifting (dashed curve) the atomic distribution in Fig. 8 to agree with the molecular one at higher transversal energies. This is because only higher transverse energies ions have high probability of undergo close encounter events that yields x-ray emission and backscattering. The same procedure was carried on for B^+ and B_2^+ (not shown).

By using the described simulation procedure, we have found the following values for Coulomb heating: 2.5 ± 0.2 eV for H_2^+ and, for the heavier molecules, the results are shown in Figure 6, where it is possible to compare with the experimental results. An observation of figure 6 shows that the theoretical curve follows the tendency of the experimental results, being the agreement, within the experimental errors, quite reasonable.

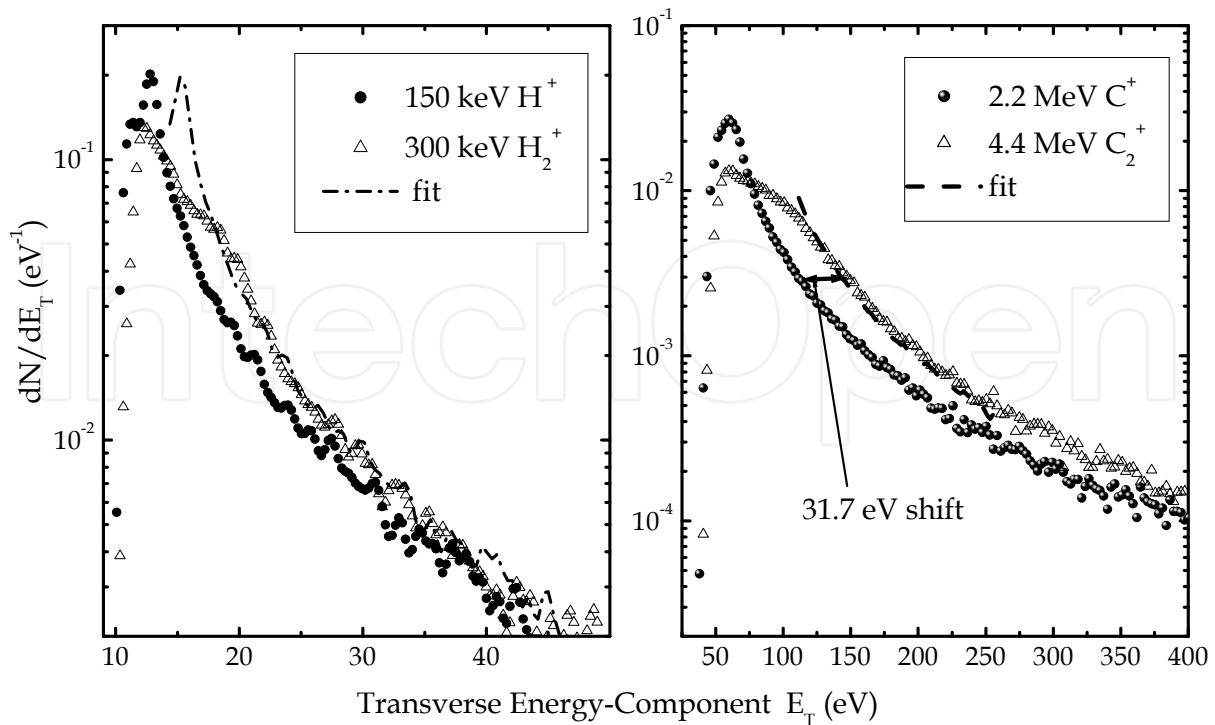


Fig. 8. Simulated transversal-energy distribution for 150 keV/atom H and H₂ (left) and 2.2 MeV/atom C and C₂ beams (right). Energy shifts are necessary in order to match the molecular and atomic beams distributions at transverse energies above 25 eV (H₂) and 150 eV (C₂) (Fadanelli et al., 2004, 2008).

5. Discussions

Since both experimental and simulated results indicate an increase of the Coulomb heating values as a function of the energy, an attempt was made in order to check whether a more general behaviour could be drawn from these data. Then, an approach was devised: instead of plotting the Coulomb heating results as a function of the ion kind and energy, we calculated the values of the potential energy per ion of the molecule in order to compare to the Coulomb heating results. To this end, we have used the DHFS calculations interpolated to the mean charge value of the ions. In Fig. 9, (Fadanelli et al., 2010) the Coulomb heating values, obtained from all the described experiments are plotted together as a function of the stored potential energy. All results lay in a single straight line, indicating some kind of “universal behaviour” where the Coulomb heating produced by the break-up of the molecules is a linear function of their potential energy per ion. In addition, the average Coulomb heating is given by:

$$\frac{1}{4\pi} \int_{\Omega} (U_0 \sin^2 \theta) \sin \theta d\theta d\phi = \frac{2U_0}{3} \quad (4)$$

where U_0 is the stored potential energy per ion and θ is the angle between the molecule axis and the initial molecule velocity.

Thus, assuming an isotropic Coulomb explosion, a plot of the Coulomb heating as a function of U_0 should be well fitted by a straight line bearing a slope of approximately 0.67. The fitted

line in Fig. 9 has a slope of 0.60 ± 0.02 , which is compatible to this prediction, namely 0.67. The difference could be attributed to second-order effects, e.g. the wake potential (Arista, 2000), not taken into account in our simulations. Therefore, the slope of the straight line of Fig. 9 indicate that two third of the Coulomb explosion energy produces the Coulomb heating. The remaining one-third gives place to the longitudinal explosion of the molecules and therefore do not contribute to the Coulomb heating, contributing to the beam straggling instead (Fadanelli et al., 2006).

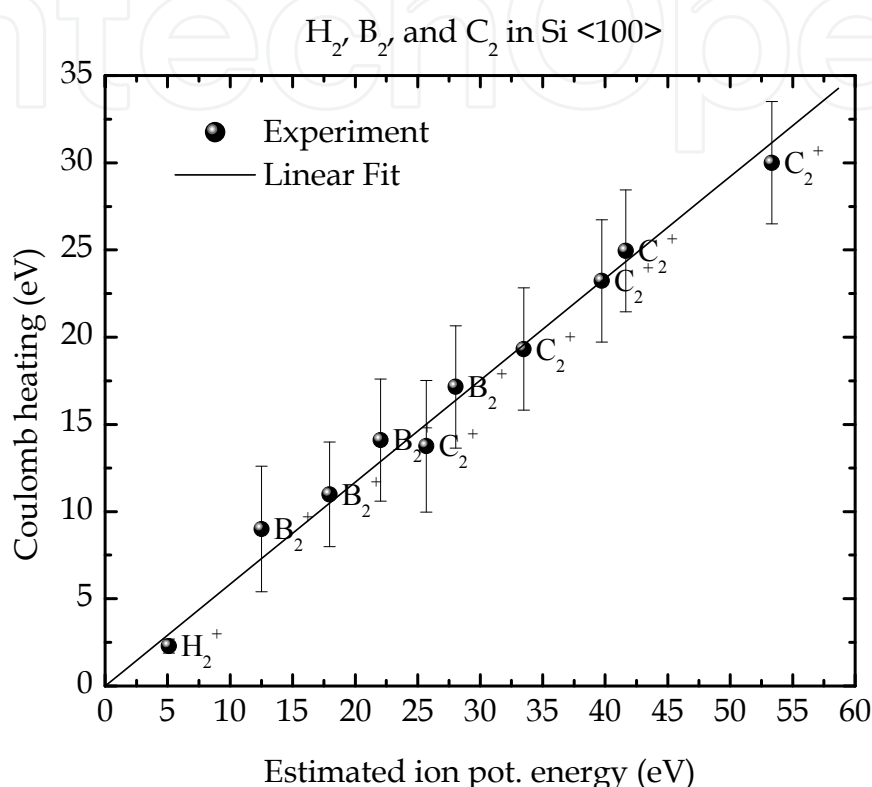


Fig. 9. Experimental Coulomb heating of H_2^+ , B_2^+ and C_2^+ projectiles as a function of the estimated DHFS screened potential energy between the projectile components. All data seems to lay in a single straight line, with a slope about 0.6. The slope seems compatible with the expected $2/3$ value (Fadanelli et al., 2010).

6. Conclusion

In summary, the present work shows that by combining monoatomic and molecular channelling RBS measurements with x-ray yields obtained by a scan around the Si <100> direction we were able to obtain the diatomic beams Coulomb heating values. Theoretical calculations are in good agreement with the experimental values. When the present results are plotted as a function of the potential energy per ion of the molecule, we have found that all the points fall on a straight line. This feature could indicate a “universal behaviour” for the Coulomb heating in the Si <100> channel. In addition, the line slope (0.6) shows compelling evidence that, at least for light molecules, the Coulomb heating corresponds to two-thirds of the stored potential energy per ion. Although this appears to be a general rule, it remains to be checked for heavier dicluster molecules travelling in other channels of Si (for instance, <110> and <111>) and other crystals.

7. Acknowledgment

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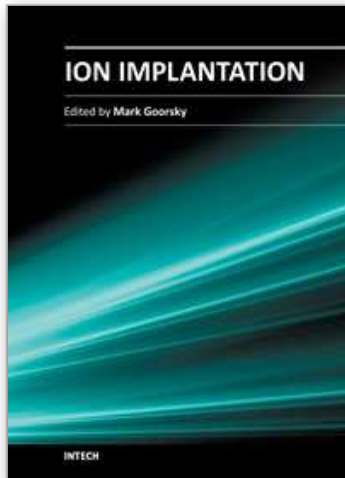
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