PHYSICAL REVIEW B **81**, 132101 (2010)

Coulomb heating behavior of fast light diclusters

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(Received 13 July 2009; revised manuscript received 30 September 2009; published 9 April 2010)

In this Brief Report we present data on the Coulomb heating induced by B_2^+ molecular beams channeling along Si⟨100⟩ direction in a large energy interval (800 up to 2200 keV/atom). The simultaneous detection of Si $K\alpha$ x rays and backscattered particles established the necessary grounds for the calculation of the Coulomb heating, i.e., the molecular transversal energy due to the Coulomb explosion transferred to the target atoms. By combining the present results with those previously published for H_2^+ at 150 keV/atom and for C_2^+ at energies from 800 up to 2200 keV/atom, we were able to obtain two striking results. First, all the experimental values fall on a straight line when they are plotted as a function of the stored potential energy per ion, suggesting some kind of "universal behavior." Second, the analysis of the whole set of experimental data shows that the Coulomb heating scales with 2/3 of the stored potential energy per ion, regardless the ion atomic number. This result is in agreement with theoretical predictions.

DOI: 10.1103/PhysRevB.81.132101 PACS number(s): 61.85.+p, 34.50.Bw, 36.40.−c

I. INTRODUCTION

It is well known that the interaction mechanism of molecular beams with matter clearly deviates from that related to the individual components of the molecule. As an example, we can mention the vicinage effect, where the molecular stopping power is larger than that corresponding to the sum of the individual components, thus indicating the existence of coherent effects.[1,2] In the case of crystalline materials, ions entering parallel to a particular axis or plane become channeled as their motion is guided by correlated collisions with target atoms. Moreover, molecules undergo a breakup process since they lose their bonding electrons due to ionization in the first monolayers of the crystal. The breakup and subsequent mutual nuclear repulsion is known as Coulomb explosion.[3] The combination of the molecular channeling and Coulomb explosion leads to the so-called transverse Coulomb heating, i.e., the transversal molecular energy transferred to the target atoms due to the Coulomb explosion. First studies of the Coulomb heating using the transmission technique were reported in early 1970s.[3] Furthermore, by studying of the behavior of H^+, H_2^+, and H_3^+ beams in backscattering geometry, Tombrello and coworkers were able to deduce the dynamics of the molecular breakup process during channeling experiments in Si crystals[4] but no attempt was done to quantify the Coulomb heating. Only much later the Coulomb heating was at last estimated from dechanneling profiles of hydrogen molecules.[5] However, the computer simulations employed in such study strongly overestimated the experimental results.

With a more sophisticated approach, Fadanelli *et al.*[6] obtained, the value of the Coulomb heating by using a combined technique, namely, the Rutherford backscattering spectrometry (RBS) with the simultaneous detection of x rays induced by H^+, H_2^+, and H_3^+ molecules at 150 keV/amu traveling around the ⟨100⟩Si channel. Moreover, the experimental values were in quite good agreement with the theoretical predictions obtained by advanced computer simulation. More recently, the same group has studied the Coulomb heating produced by C^+ and C_2^+ molecules in a wide energy range with a twofold purpose: first, to observe the behavior of the Coulomb heating as a function of the energy; and second, to test if the theoretical model employed for the simple case of hydrogen molecules could be applied to more complex ones. The results of this work[7] have shown that the Coulomb heating values increase with the beam energy in a nonlinear way. Moreover, the theoretical-experimental agreement was rather good, proving that the theory was suitable for heavier diclusters.

This Brief Report presents the quantitative determination of the Coulomb heating of B_2^+ molecules channeled around the Si⟨100⟩ direction spanning an energy interval between 800 and 2200 keV/amu. The main purpose of the present work is to draw a more general relationship of the Coulomb heating values based on the results obtained from H_2^+, C_2^+, and B_2^+ molecules interacting with crystal silicon. To this end, we have used the same experimental setup as described in detail in previous publications.[6,7]

II. EXPERIMENT AND DATA ANALYSIS

The Si⟨100⟩ samples were mounted on a four axis goniometer for the channeling procedure (details in Ref. 7). The Si⟨100⟩ channel was determined by using a 1.2 MeV alpha beam in order to minimize the radiation damage induced by the B^+ and B_2^+ beams on the target. The backscattered particles were detected by a Si surface barrier detector located at 170° with respect to the incoming beam. The overall resolution of the RBS system was around 30 keV for boron ions. On the other hand, the 1.74 keV x rays emitted from the target were detected by a Si(Li) detector with an energy resolution of 190 eV at 5.9 keV. This detector was mounted at 135° with respect to the beam direction.

Once the Si⟨100⟩ channel was determined, the sample position was changed in order to get a pristine spot free of damage caused by the α beam. Subsequently, we have changed to a B^+ beam and performed an angular scan around the Si⟨100⟩ direction from −3° up to +3° in steps of 0.2°. At each step the 1.74 keV x-ray yield and the corresponding RBS spectrum were simultaneously recorded. Upon completion of the scanning procedure, the target position was changed to a fresh spot and then the same steps were carried out with a B_2^+ beam. For each beam energy, at least five independent measurements of the x-ray yields and the corresponding RBS spectra were obtained. Therefore, the uncertainties quoted for the final results were calculated taking into account these independent measurements as well as the fittings to the spectra.

The results of the present measurements display two major features. First, the results of the x-ray emission induced by the B^+ and B_2^+ molecules show that at large angles (corresponding to the random direction) the yield induced by both beams are equal within the experimental uncertainties. However, at zero or near zero polar angles the x-ray yield induced by the B_2^+ molecule is larger (on the order of 15%) than the corresponding one produced by the B^+ beam. This difference is statistically significant and occurs for all the investigated energies. Second, the RBS dechanneling profiles observed for the B_2^+ beam is always larger than those related to the B^+ beam. Again, this is a common feature to all energies studied in the present work. Both results are clear signatures of the Coulomb explosion, which provides an extra transversal energy to the ions, thus enhancing x-ray and dechanneling yields.

In order to obtain quantitative results for the Coulomb heating it is necessary to find the dechanneling rate α as a function of the tilt angle ψ. To this end, we have used the two beam model[8,9] which has been employed successfully in previous publications.[6,7] In brief, this model assumes that the ion beam can be divided in two parts: the channeled and dechanneled fractions and their relation is given by

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

where α is the dechanneling rate, $\chi(z)$ is the dechanneled fraction at a depth z inside the channel and $[1-\chi(z)]$ is the channeled fraction which is a function of the penetration depth as well. The α factor is assumed constant and its values is determined from the fitting of the corresponding atomic and molecular RBS spectra. In order to obtain good fittings of the RBS spectra, it has been shown[6,7] that we must take into account that the effect of the Coulomb explosion on the ion flux occurs only after a certain time delay which, in turn, corresponds to a delay distance (denoted as Δ_z) traveled inside the crystal. Indeed, the molecular breakup begins as soon as the B_2^+ molecular ions penetrate a few monolayers within the target, while the Coulomb explosion usually takes several hundreds of angstroms of ion penetration to change the shape of the ion flux and, consequently, the transverse energy distribution. Although this is a continuous process, we assume that all the changes in the ion flux occur after a distance Δ_z. In the case of the H_2^+, Δ_z was on the order of 40 nm at 150 keV/amu, while for C_2^+ its value varied from 120 nm for the lowest energy up to 220 nm for the highest one. Consequently, the fits of the RBS molecular spectra obtained for the boron case were done with two free parameters: $\alpha(\psi)$ and Δ_z. The fitting procedure yields several $[\alpha(\psi),\Delta_z]$ pairs which correspond to a reasonable fit of the B_2^+ spectra. Then, as described in detail in Ref. 7, we have fitted the corresponding x-ray yield as well, obtaining an unique pair of values for $\alpha(\psi)$ and Δ_z. For the boron case, Δ_z values varied from 30 nm for the lowest investigated energy up to 120 nm for the highest one.

III. RESULTS AND DISCUSSION

Once all $\alpha(\psi)$ are known, we can find the Coulomb heating energy through the Hamiltonian of the transverse ion motion which, for small ψ angles, reads

$$H_{\perp} = E_0\psi^2 + V(r), \tag{2}$$

where E_0 is the beam energy per atom and $V(r)$ is the channel potential. As discussed above, the molecular breakup process starts as soon as the B_2^+ molecule penetrates a few monolayers in the target. We estimated that this time (about 10^{-15} s) is much shorter than the typical time (10^{-14} s) for a single particle to get channeled. In this scenario, we can assume that the Coulomb explosion acts like as an initial divergence or "astigmatic lens"[8] and increases the initial transversal energy by an effective Coulomb heating energy ΔE_C. The increase in the transversal energy has a direct impact on the dechanneling rate $\alpha(\psi)$. Since the initial transversal energy $H_{\perp}$ depends on the incident angle (through the Hamiltonian), the additional Coulomb heating energy component ΔE_C will be equivalent to a tilt offset $\Psi \rightarrow \sqrt{\Psi + \Delta E_C/E_0}$. It has been shown[7] that the parameterization of the atomic dechanneling rate $\alpha_{atom}(\psi)$ leads to a simple relation between the molecular dechanneling rate $\alpha_{mol}(\psi{=}0)$ and the atomic dechanneling rate,

$$\alpha_{mol}(\psi = 0) = \alpha_{atom}(\sqrt{\Delta E_C/E_0}). \tag{3}$$

Therefore, we can easily deduce from the above expression that the relationship between the tilt offset $\Delta\psi$ and the Coulomb heating energy ΔE_C is given by

$$\Delta\Psi = \sqrt{\Delta E_C/E_0}. \tag{4}$$

In this way we were able to obtain the Coulomb heating values corresponding to each energy. For further details and considerations we refer the reader to Ref. 7. The experimental values of the Coulomb heating as a function of the energy are displayed in Fig. 1. As can be observed, they vary from 9 eV at 0.8 MeV/atom up to 17 eV at 2.2 MeV/atom.

A Monte Carlo simulation was developed in order to determine theoretically the channeling of the B^+ and B_2^+ ions along the Si⟨100⟩ channel. To describe the ions motions in a simple way, several assumptions were used. The Si⟨100⟩ channel is described using the continuous potential model[9–11] and the motion of the ionic ensembles were described classically. Since the ion charge state is supposed to reach equilibrium after traversing the first target layers, the charge states were chosen randomly based on a Gaussian distribution.[12,13] The chosen charge state is kept constant throughout the entire ion path. The interaction between the two ionic components of the molecule is modeled using an

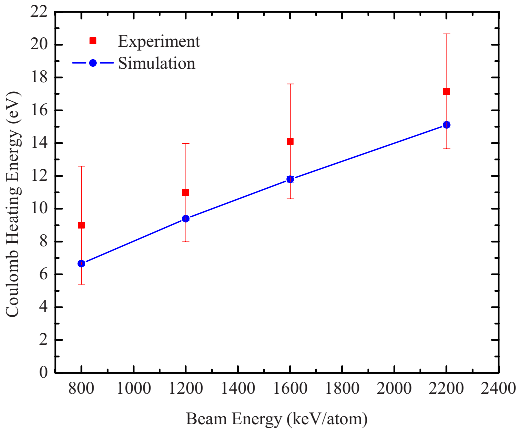

FIG. 1. (Color online) Coulomb heating energy produced by B_2^+ molecules channeling around the Si⟨100⟩ direction as a function of the beam energy. The square symbols stand for the experimental values and the circles represent the results obtained by the simulation described in the text. The line is drawn to guide the eye.

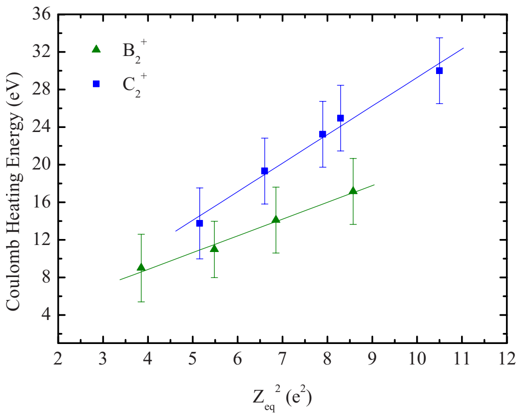

FIG. 2. (Color online) Coulomb heating energy produced by B_2^+ (triangles) and C_2^+ (squares) molecules channeling around the Si⟨100⟩ direction as a function of the square of the equilibrium charge Z_{eq}. The C_2^+ data were taken from Ref. 7. The straight lines are linear fittings to each data set.

analytical fitting to the Dirac-Hartree-Fock-Slater (DHFS) calculations for the screened ion potential.[14,15] This was done taking into account the chosen charge state for each B^+ ion in the target. Moreover, we have assumed an initial ion separation of 4.164 a.u.[16] The simulation output is the transverse kinetic-energy distribution for each ion (B^+ and B_2^+) ensemble. By shifting the B^+ distribution until it matches the B_2^+ one for medium transverse energies, it is possible to estimate the theoretical Coulomb heating values.[7] As can be observed in Fig. 1, there is, within the uncertainties, a good agreement between theory and experiment. The theoretical predictions follow the same trend as the experimental results. However, they seem to underestimate systematically the experimental ones. This can be attributed to the simplifications done in the model (see Ref. 7 for details).

Taking into account that the present results as well as the ones published previously for C_2^+ seems to suggest an increase in the Coulomb heating values as a function of the energy, a first attempt was made in order to check whether a more general behavior could be drawn from these data. To that end we have plotted, for each ion and beam energy, the experimental values as a function of the square of the estimated equilibrium charge state Z_{eq}. This was done because the Coulomb explosion, in a first approximation, should depend on the repulsion between the components of the molecule, which is characterized by the Z_{eq}^2. These results are displayed in Fig. 2. As can be observed, the behavior of the Coulomb heating for both molecules follows a linear relationship as a function of Z_{eq}^2 of each molecule. However, despite the fact that we have obtained a linear relationship, this behavior differs for B_2^+ and C_2^+ molecules.

A different approach was then devised where the rather simplistic procedure adopted above was replaced by the calculation of the internal potential energy per ion stored in the molecule traveling inside the crystal. To this end, we have used the DHFS calculations interpolated to the mean charge value of the ions. In Fig. 3, the Coulomb heating values obtained from the present experiment are plotted together with the ones corresponding to H_2^+ (Ref. 6) and C_2^+ (Ref. 7) and as a function of the internal potential energy per ion of the molecule. All results lay in a single straight line, indicating some kind of "universal behavior" where the Coulomb heating produced by the breakup of the molecules is a linear function of their potential energy per ion. In addition, as discussed in Ref. 6, the average Coulomb heating given by

$$\frac{1}{4\pi}\int_{\Omega}(U_0 \sin^2 \theta)\sin \theta d\theta d\phi = \frac{2U_0}{3}, \tag{5}$$

where U_0 is the stored potential energy per ion and θ is the angle between the molecule axis and the initial molecule velocity. This result indicates that only 2/3 of the stored energy in the molecule contributed to the Coulomb heating. The remaining one third goes to the longitudinal explosion of the molecules and therefore do not contributed to the Coulomb heating. The fitted line in Fig. 3 has a slope of

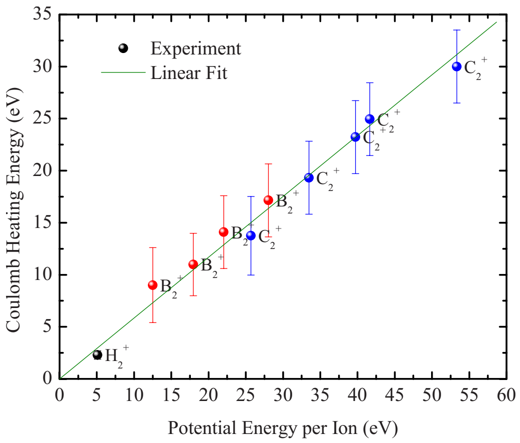

FIG. 3. (Color online) Coulomb heating energy produced by H_2^+, B_2^+, and C_2^+ molecules channeling around the Si⟨100⟩ direction as a function of the internal stored potential energy of the molecule per ion. The H_2^+ and C_2^+ data were taken from 6 and 7, respectively. The straight line is a linear fitting to all data.

0.60 ± 0.02, which is in good agreement with the theoretical prediction of Eq. (5), namely, 0.67.

IV. CONCLUDING REMARKS

In summary, the present work shows that the simultaneous measurement of RBS spectra and x rays induced by B^+ and B_2^+ ions channeling around the Si⟨100⟩ direction allowed us to obtain the B_2^+ Coulomb heating energy at several energies. The Coulomb heating values vary from 9 eV at 800 keV/atom up to 17 eV at 2200 keV/atom. Theoretical calculations are in good agreement with the experimental values. When the present results are plotted with those obtained previously for H_2^+ and C_2^+ molecules as a function of the stored potential energy per ion of the molecule, we have found that all the points fall on a straight line. This feature could indicate an universal behavior for the Coulomb heating in the Si⟨100⟩ channel. In addition, the slope of the straight line (0.6) can be taken as a compelling evidence that, at least for light molecules, the Coulomb heating corresponds to two thirds of the stored potential energy per ion as predicted by the theory. Although this behavior appears to be a general rule, it remains to be checked if it holds for other Si channels and/or for heavier diclusters molecules.

ACKNOWLEDGMENT

The authors are indebted to the Brazilian agency CNPq (Conselho Nacional de Desenvolvimento Científico e Tecnológico) for the support of this work.

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