Experimental energy loss of slow H^+ and H_2^+ in channeling conditions

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The interactions of hydrogen molecular ions (H_2^+) and protons in the low energy range ($E<7.5$ keV/u) with very thin foils of monocrystalline gold are experimentally studied. Measurements of energy loss distributions of molecular fragments, recombined molecules, and protons channeled in the $\langle 100 \rangle$ direction of a monocrystalline gold thin film have been performed. From the energy loss data we determine the stopping power ratio, which shows a significant negative ''vicinage effect,'' giving a reduced energy loss of molecular fragments and recombined molecules as compared to the energy loss of protons. This effect is more pronounced in the case of detected molecules. Computational simulations, where nonlinear models are included, qualitatively agree with our experimental results and suggest that some pairs of ions travel and emerge from the crystal with appropriate internuclear distances and relative velocities and are able to recombine at the exit.

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The study of the interaction of ions with monocrystalline and polycrystalline solids is one of the most active areas of physics and a source of multiple technological applications. When the target is monocrystalline, the situation is special because the distribution of impact parameters and the yields of the physical processes are found to be very dependent on the relative orientations of the beam and target [1] giving rise to the phenomenon of channeling. This effect has been intensively studied, and is currently used as a tool in ion implantation studies and ion-beam modification of materials.

The special conditions in the channeling process (the lack of homogeneity of the electron density, the ordered distributions of atoms, etc.) make of channeling an important tool for testing energy loss theories, electron band theories, and interatomic potentials [1–5].

When the projectile is a molecule *the situation is specially* interesting because the closeness of the projectile components makes the interaction with the target nuclei and electrons rather different from that of the single ion beam. This so called ''vicinage effect'' allows us to obtain worthy information on dynamical effects in solids, including elastic and inelastic collisions, screening effect, and interactions with collective modes of the material [6]. The determination of the ratios between the energy loss of the molecular fragments and that of individual protons gives a measurement of these ''vicinage effects.'' This problem has been previously theoretically and experimentally addressed, mainly in the high and medium energy range ($E \geqslant E_F$) where energy loss ratios higher than 1 were found [7]. Most of these studies were made using amorphous or polycrystalline solids, but a theoretical prediction of important vicinage effects for correlated protons in channeling conditions was made long ago [8].

Here, an experimental and theoretically study of the energy loss of transmitted H_2^+ and H^+ fragments along the $\langle 100 \rangle$ channel of gold in the low energy regime is presented, extending for the first time the studied energy range down to 1 keV/u.

The energy loss measurements for protons, molecular fragments of dissociated hydrogen molecules, and hydrogen molecules from recombined molecular fragments, were performed using the transmission geometry after traversing very thin monocrystalline foils of Au. We detected the emerging beam within a very small angular cone ($<0.8°$) along the incident beam direction. All these measurements were performed in the energy range from 1 to 7.5 keV/u. In our experimental setup, the beam from a hot discharge ion source (Colutron Research Corp., Boulder, CO, USA) was focused

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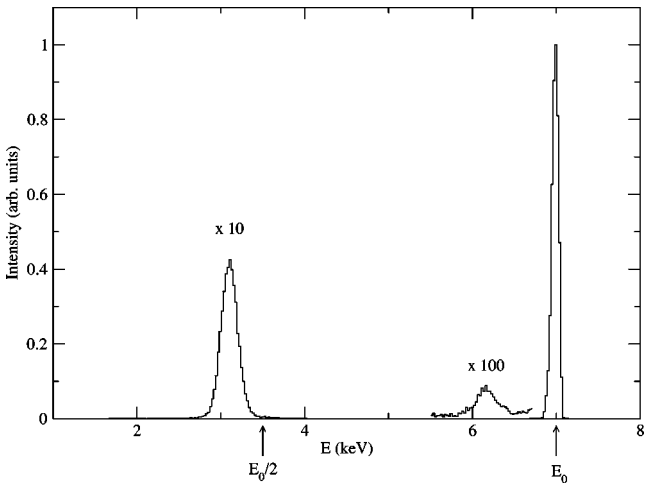

FIG. 1. Energy spectrum corresponding to 3.5 keV/u incident H_2^+ beam. The intensity of the molecular fragments peak is multiplied by 10 whereas the detected molecules (recombined fragments) peak is multiplied by 100. Also, the spectrum shows a peak of molecules transmitted through a pinhole with no energy loss.

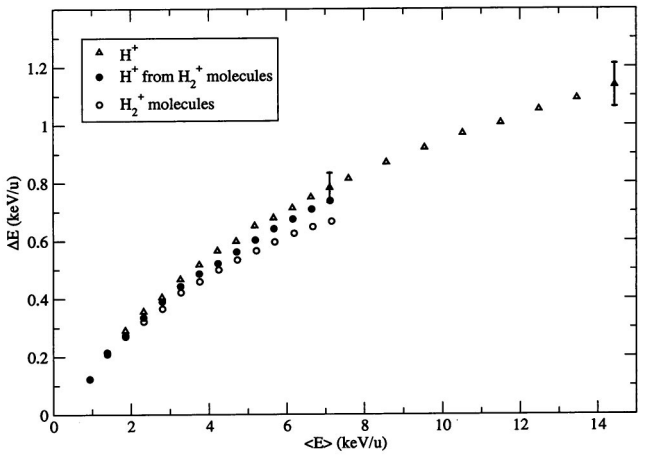

FIG. 2. Energy loss per unit mass as a function of the mean energy inside the film as defined previously. The open triangles correspond to the energy loss of emerging protons with incident energy from 2 to 15 keV, full circles correspond to molecular fragments from the H_2^+ dissociation, and the open circles correspond to the detected recombined fragments (molecules).

by an electrostatic lens system and mass analyzed by a velocity filter. The proton energy analysis was performed by a 160° spherical electrostatic analyzer (Comstock Inc., Oak Ridge, TN, USA) of less than 1% of resolution. The associated error in the determination of ion energy was less than 3%. Ions were detected with a microchannel plate system in chevron configuration. All the system was evacuated by two turbo molecular pumps and a liquid nitrogen trapped oil diffusion pump. The operating pressure was less than 2×10^{-7} Torr. The monocrystalline gold targets were of commercial type (Ted Pella Inc., Redding, CA, USA), these specimens provide a valuable test for electronic microscopes. The gold is evaporated onto a Cu grid and induced to grow in a (100) orientation. According to the manufacturer's specifications, the films thicknesses ranged from 9 to 11 nm. The target was mounted on a five axis goniometer so that the film surface was nearly perpendicular to the beam with a precision better than 0.2°.

The accepted physical picture when a H_2^+ molecule collide with a metallic medium indicates that when it penetrates the solid it loses the binding electron in the first atomic planes [9]. Consequently the resulting fragments move in a correlated way through the solid. Eventually the leaving fragments can form a bound state at the exit if special conditions are fulfilled [10]. Therefore, when bombarding the foil with H_2^+ molecules one should obtain at the exit an intense peak corresponding to the fragments of the dissociated molecules and a small peak corresponding to molecules recombined at the exit surface. The dwell time in our experiment (≈ 8 fsec.) almost completely reduce the possibility of a direct transmission regime for molecules which is observed at high energies [10]. In Fig. 1, we present a typical energy spectrum corresponding to the case of a 3.5 keV/u H_2^+ beam incident in a Au $\langle 100 \rangle$ film. One clearly sees a peak at the incident energy (E_0) corresponding to a fraction of molecules going through target pinholes with no energy loss. Another peak near one half the incident energy corresponds to slowed down molecular fragments traveling across the film and a smaller peak close to the incident peak corresponds to the recombined fragments. Figure 2 shows a set of measured energy loss data in keV/u as a function of the mean energy of projectile inside the solid $\langle E \rangle$, defined by $\sqrt{\langle E \rangle} = \frac{1}{2}(\sqrt{E_0} + \sqrt{E_1})$ (velocity average), where E_0 is the mean incident energy and E_1 is the mean value of the exit-energy distribution. In order to compare the different energy losses of protons, fragments, and molecules we present the plot as energy loss per unit mass. The energy loss of the molecular fragments and recombined molecules (per unit mass) are undoubtedly smaller than the energy loss of protons at the same energy per unit mass (i.e., at the same velocity). In Fig. 3, we show the energy loss divided by the square root of mean energy, $\langle E \rangle^{1/2}$, as a function of $\langle E \rangle^{1/2}$. This representation

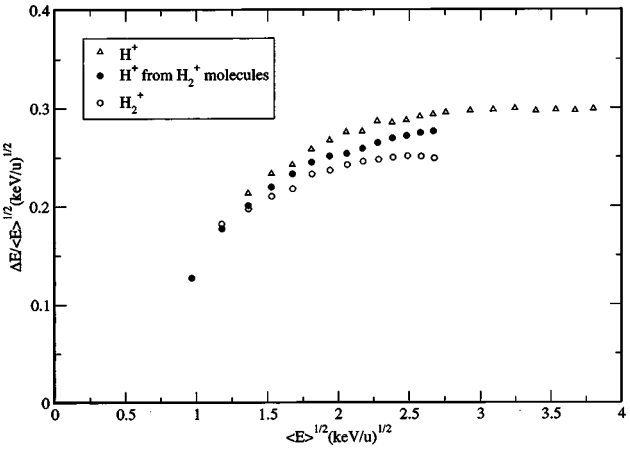

FIG. 3. Energy loss per unit mass divided by the square root of the mean energy, $\langle E \rangle^{1/2}$, as a function of $\langle E \rangle^{1/2}$. Data points are the same as Fig. 2. This graph shows the threshold effect in the energy loss, i.e., departure from the velocity proportionality of the stopping power for the three type of particles.

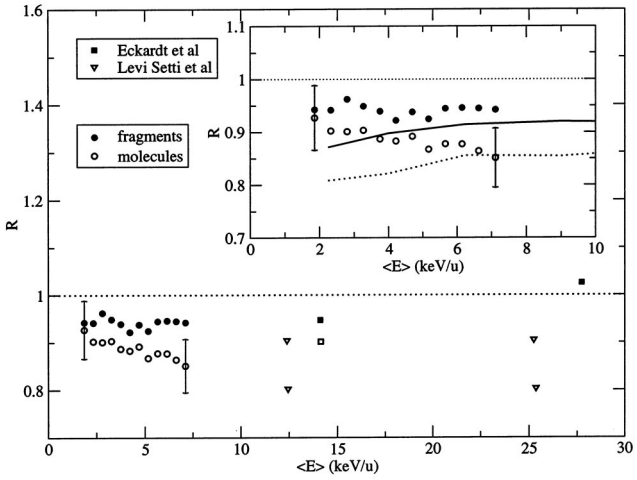

FIG. 4. Energy loss ratio R for molecular fragments (full symbols) and recombined molecules (open symbols). The circles represent our experimental data while other symbols represent experiments from other authors for amorphous carbon foils. In the inset we observe a detail of our experimental results compared with our computational simulations. The solid line corresponds to the fragments ratio and dashed line is the molecular ratio.

should yield a constant value if the energy loss were proportional to the projectile velocity. These data show the typical ''threshold effect'' (or band-structure effect) present in the energy loss in transition metals at this energy range which has been reported elsewhere [11,12] and has been associated to the excitation of *d*-band electrons. In this graph we also observe for first time the threshold effect for molecular fragments and molecules.

In Fig. 4, we present the energy loss ratio R, defined as $\Delta E_{\mathrm{frag}}/\Delta E_{\mathrm{H}^+}$ in the case of individual molecular fragments and $\Delta E_{\mathrm{H}_2{}^+}/2\Delta E_{\mathrm{H}^+}$ in the case of recombined molecules, as a function of the incident energy per unit mass. Values of R significantly different from 1 imply the presence of vicinage effects in the energy loss. The circles depict our experimental results for Au $\langle 100 \rangle$ and the squares and triangles represent experimental results for amorphous carbon films from other authors [13,14]. Full symbols indicated the ratio for molecular fragments while open symbols represent the case of recombined molecules. We observe values of R significantly lower than 1. This tendency has also been found in other experiment in this energy range [13,14], although for other targets and not in channeling experiments. On the other hand an opposite behavior has been observed at higher energies ($E > E_F$) where values of R larger than 1 have been found [7].

In order to analyze these results and gain physical insight we have performed computer simulations. The basis of these simulations for the case of atomic ions have been reported before [3–5]. Here the simulation code has been extended to treat molecular fragments according to the following: we assume that the molecule dissociates as soon as it enters the foils. Then the resulting fragments trajectories are described classically and controlled by the influence of three forces: the interaction with the fcc Au atoms modeled by a Molière potential [15]; the stopping force due to electronic excitation modeled by nonlinear theory [16] and a local electronic density within the channel calculated by the method linear muffin-tin orbitals [17,18], and finally the screened Coulomb repulsion between the fragments was modeled by a Yukawa potential [7]. To account for the vicinage effect in the energy loss, we have included in the stopping process the nonlinear vicinage effects using the results of Diez-Muiño and Salin [19] for a hydrogenic dimer in an electron gas at low velocities. At the exit of the foil we determine whether the fragments can form a bound state depending on the distribution of distances between the fragments, their relative energies, and the potential-energy curve of the ground state of the $H_2{}^+$ molecule [20].

In the inset of Fig. 4, we can see a detail of our experimental data compared with computer simulations results. The full line corresponds to the case of fragments while the dotted line represents the ratio for recombined molecules. The simulations qualitatively agree and show the same trend that the experiments, i.e., values of R lower than 1 for molecular fragments and an even lower ratio for recombined molecules. The reason why the recombined molecules have lower values of R is that according to our simulations a condition that enhances the possibility of recombination is that the fragments do not separate too much inside the foil, and the vicinage effect in the relevant range of internuclear distances is less than one.

In summary, a comparative study of channeling at very low energies using protons and $H_2{}^+$ ions was performed. Transmission of $H_2{}^+$ molecules was observed and the energy losses of molecular fragments as well as transmitted $H_2{}^+$ ions under channeling conditions were determined in this very low energy range. The experiment shows unambiguously a diminished energy loss for molecular fragments and recombined molecules as compared to individual protons ($R<1$). The lower energy loss per unit mass for recombined molecules observed here is explained by computer simulations that show that the fragments that recombine at the exit surface are predominantly those that remained relatively close inside the foil. In addition, we find that our model (based on nonlinear calculations for hydrogenic dimers [19]) describes reasonably well the behavior of the stopping ratio in monocrystalline targets.

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