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Recombination of H_2^+ molecules in channeling conditions

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Abstract

The transmission of H_2^+ molecules through thin gold foils in channeling conditions is studied, both theoretically and experimentally, in the low energy range ($E < 10$ keV/u). For this purpose we have performed semiclassical computer simulations of the fragment trajectories inside the target and we are able to determine if they can reconstruct at the exit surface via the recombination model. Our calculated H_2^+ transmission yield shows interesting oscillations as a function of the dwell time, and also the calculated exiting number of H_2^+ with respect to H^+ shows a very good agreement with our experimental measurements. We observed transmission yields almost three orders of magnitude larger than in the case of a random target.
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1. Introduction

The interaction of molecular beams with solids has been studied thoroughly in the past, both theoretically and experimentally, because it provides valuable information about the basic mechanisms of solid response to perturbations [1]. When the projectile is incident along a crystalline direction of a monocrystalline solid, the situation is rather different than when the same projectile is interacting with a random solid because the distributions of impact parameters are different in both cases, giving rise to the phenomenon of "channeling" [2]. As an example of this behaviour, it is found that the energy losses and ranges in channeling conditions differ significantly from the random case [2–4]. Also, the "vicinage effects" in the energy loss of molecular beams were found to be different in channeling conditions [5]. In this work we will focus on the influence of channeling in the transmission of H_2^+ molecules through thin foils.

This problem was previously studied experimentally by Poizat and Remillieux [6] and an enhancement of the transmission yield through planar directions of the crystal was found. Also, several papers have studied the transmission of molecules in random solids [7–11]. Here we will study, both theoretically and experimentally, the transmission of H_2^+ molecules along several crystalline directions of a Au film in the low energy range ($E < 10$ keV/u) and make a comparison with a similar situation in a random target. For this purpose, we have developed a numerical code that follows the molecular fragments trajectories inside the monocrystalline solid and then modelled the probability of recombination at the exit of the foil.

In what follows, we present the details of our simulation code and the model to evaluate the probability of molecular recombination (Section 2). The main results are discussed in Section 3 and the conclusions appear in Section 4.

2. Theoretical and simulation background

When a H_2^+ molecule penetrates a solid target it can be transmitted without dissociation if its dwell time inside the foil is very small, typically lower than 0.2 fs [7,8]. This situation cannot be fulfilled at low molecular energies ($E < 10$ keV/u) but at higher energies and very thin foils we can find at the exit molecules that are transmitted without dissociation [8]. Nevertheless H_2^+ can still be transmitted if the molecules dissociates, one of fragments captures a target electron, and they recombine at the exit into a molecular bound state. Therefore, we can say that for our range of interest (dwell times ≈ 10 fs.), the H_2^+ molecule dissociates as soon as it enters the foil and that the process of recombination is responsible for all the molecules obtained at the exit. Once inside the solid, the dissociation fragments move in a correlated way and so their separation R and the relative kinetic energy E_{rel} are affected by several interactions.

In order to describe the trajectories of the dissociation fragments throughout the monocrystalline gold foils, we have used a simulation code described previously [4,12] that solves the classical equations of motion of the resulting fragments

and contains the main dynamical interactions with the target nuclei and electrons and also with the molecular partner. The multiple scattering with the atomic cores of the solid is mainly responsible for the fragments trajectories and was modelled by a repulsive Molière potential [13]. We include atomic vibrations of the solid allowing the crystal atoms to displace from their equilibrium positions through the Debye model [2]. The energy loss of the fragments is mainly due to the target electronic excitation and was included in the simulation code making the fragments lose energy as if they were in a jellium with a density corresponding to the local valence electronic density. The stopping model is non-linear [14] also including energy loss straggling [15] and the local electronic density in the primitive cell was calculated using the TB-LMTO method [16,17]. The screened Coulombic repulsion with the molecular partner was modelled by a Yukawa potential [1]. The initial conditions of incidence of the H_2^+ molecule are as follows: random orientation of the internuclear axis, an initial internuclear distance of approximately 1.25 Å [18], random impact parameter across the channel and velocity directed along a target crystalline direction.

The model we have used to investigate the recombination of H_2^+ molecules at the exit of the foil has been used before [8,10,11] and assumes the following sequence of events: (a) when interacting with the solid, the fragments can capture or lose electrons. At the exit of the foil, the fragments charge states can be H^0, H^+ or H^- with probabilities depending on the velocity of the fragments and to a lesser extent on the target material. These probabilities were approximated by the charge fractions ($\phi^0 + \phi^+ + \phi^- = 1$) obtained experimentally in amorphous gold [19]. (b) If H^+ and H^0 belonging to the same molecular ion meet at the rear surface with a given internuclear distance R and a relative kinetic energy $E_{rel} = \frac{1}{2}\mu\dot{R}^2$ recombination into a molecular bound state can take place (μ is the reduced mass of the molecule). This will occur if $U(R) + E_{rel} < 0$, being $U(r)$ the potential energy of the $1s\Sigma_g$ molecular state relative to that of the $H^0(1s) + H^+$ separated atoms which is taken to be zero [20]. We assume that the probability of the fragments to be captured

into other molecular state different than $1s\Sigma_g$ is negligible. This assumption is supported by a good comparison of the experimental number of H_2^+ molecules recombined at the exit obtained in this work and that in [11].

In our simulations, we store the values of R and E_{rel} obtained at the exit of a foil of a given thickness for all the histories, and then, taking into account the probability of H^+ and H^0 encounter, we calculate the number of H_2^+ obtained by recombination with the model just described. The number of exiting H^+ can be due to the isolated fragments leaving the foil's rear surface or due to exiting pairs that do not recombine.

3. Results

In Fig. 1, we have depicted the calculated transmission yield, i.e. the number of exiting H_2^+ molecules relative to the number of incident molecules N_{hist}, as a function of the dwell time. The projectiles are 6.25 keV/u H_2^+ molecules inciding in a monocrystalline Au foil oriented in several crystallographic directions. We observe that the transmission yield decreases with the dwell time. The main mechanism that tends to separate the fragments reducing the probability of recombination is the collision with the target nuclei. In this energy

range, the Coulombic repulsion between the fragments is so screened that it can be neglected as a mechanism of fragment separation. The scattering with the target nuclei explains the decrease of the yield with the thickness and also why the recombination is more enhanced if the crystal is oriented in the $\langle 1\,1\,0 \rangle$ direction. The $\langle 1\,1\,0 \rangle$ direction is the most open in an fcc crystal and have larger channels reducing the probability of a close encounter with a crystal atom. The channel area is $0.35a^2$ for this direction, the $\langle 1\,0\,0 \rangle$ direction has an intermediate channel size ($0.25a^2$) and the most closed surface is the one corresponding to the crystal oriented in the $\langle 1\,1\,1 \rangle$ direction ($0.144a^2$), a being the lattice parameter. We also observe interesting oscillations of the transmission yield with the dwell time. In order to interpret these oscillations, we show in Fig. 2, the calculated transmission yield as a function of the dwell time for the $\langle 1\,0\,0 \rangle$ crystal orientation but forcing the initial molecular orientation to be perpendicular or parallel respect to the channel axis. We can see that if the molecule is initially aligned with the axis much higher transmission yields are found because the scattering of the fragments with the target nuclei is thus reduced and no oscillations in the yield are present. However, if the molecule impinges with its internuclear axis perpendicular to the channel axis we can see

Fig. 1. Number of H_2^+ molecules obtained at the exit of an Au foil relative to incident number of molecules, as a function of the dwell time. The molecular energy is 6.25 keV/u and three different orientations of the crystal were included.
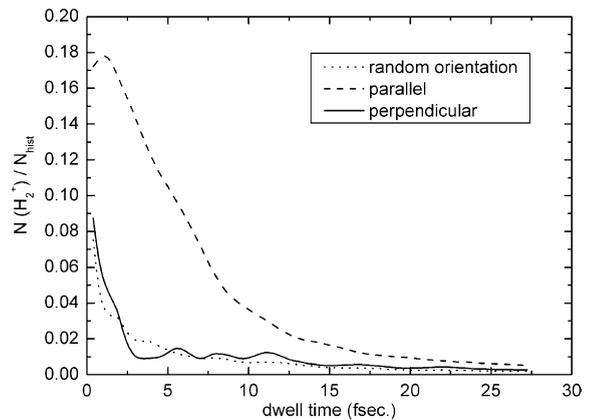
Fig. 2. Number of H_2^+ molecules obtained at the exit of the Au foil relative to the incident number of molecules, as a function of the dwell time. The crystal orientation is the $\langle 1\,0\,0 \rangle$ and the molecular axis at the entrance can be parallel, perpendicular or random with respect to the $\langle 1\,0\,0 \rangle$ direction.

much lower transmission yields and evident oscillations as a function of the thickness. In this latter case, there are histories where the fragments can travel in adjacent channels and the internuclear distance have oscillations with the target thickness giving rise to the just mentioned oscillations in the recombination yield.

Before the comparison with the experimental results it is necessary to make some comments about our experimental setup. The setup is the same we have been using for energy loss measurements and has been previously described [12]. Concisely it consists of a H_2^+ beam incident on commercial thin monocrystalline foils in transmission geometry. The emerging particles were energy analysed with an electrostatic detector. The integration of the energy loss spectrum during a fixed time gives us information about the number of particles detected. As the electrostatic analyzer only detects charged particles and we cannot normalize the peak intensities relative to the incident beam we have compared the H_2^+ relative to the H^+ peak intensity in the same exit energy spectrum in order to obtain experimental information about the recombination process. In Fig. 3 we show the number of H_2^+ relative to that of H^+ obtained at

the exit of a 110 Å thick monocrystalline gold foil for different projectile energies and three different crystal orientations. The symbols represent our experimental results for Au $\langle 1\,0\,0 \rangle$. Again we can see that the most open orientation, namely the $\langle 1\,1\,0 \rangle$, produces larger recombination rates, intermediate for the case of the $\langle 1\,0\,0 \rangle$ orientation and small recombination in the case of the $\langle 1\,1\,1 \rangle$ orientation. The agreement of the $\langle 1\,0\,0 \rangle$ simulation with the experimental data is very satisfactory. As a matter of comparison with recombination in random targets, we have added in the lower part of Fig. 3, the results corresponding to a simulation for a 110 Å thick amorphous gold foil. In these simulations we have only included a Monte-Carlo model [21,22] for the nuclear multiple scattering of the molecular fragments and the same energy loss model as in the simulations in monocrystalline solids but considering uniform electronic density. Clearly, the presence of multiple scattering with target nuclei reduces by almost three orders of magnitude the recombination yield as compared to the channeling case. This explains why, within our experimental precision, we cannot observe H_2^+ molecules at the exit of a 110 Å thick polycrystalline gold foils but we do observe H_2^+ molecules in similar monocrystalline foils.
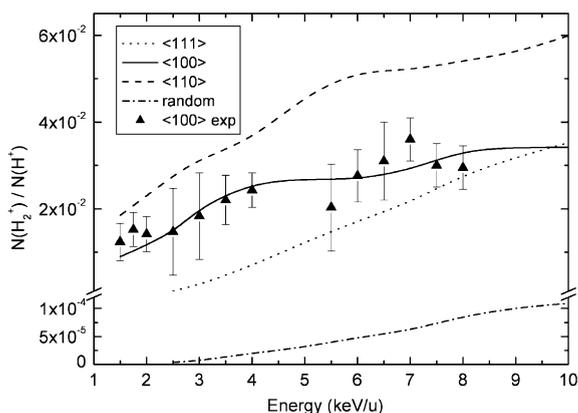
Fig. 3. Number of H_2^+ molecules compared to the number of H^+ fragments exiting a 110 Å thick Au foil as a function of the projectile energy. The lines in the upper part of the figure correspond to our simulations for three different orientations of the Au monocrystal, while the symbols are our experimental results for Au $\langle 1\,0\,0 \rangle$. The line in the lower part of the figure correspond to a simulation in a foil of amorphous Au.

4. Conclusions

The transmission of H_2^+ molecules through thin gold foils in channeling conditions has been studied experimentally and also with computer simulations.

The scattering with the target nuclei is the main responsible for the fragments separation and their relative energy when exiting the foil and so recombination is clearly enhanced in channeling conditions with respect to random targets. In the same way, the crystal oriented in directions resulting in the most open surfaces ($\langle 1\,1\,0 \rangle$ in the case of an fcc crystal) give rise to larger transmission yields.

We find interesting oscillations, not yet measured, in the recombination yield as a function of the dwell time in channeling conditions due to histories where the fragments of the H_2^+ molecule travel in adjacent channels and the intermolecular

distance presents oscillations with the target thickness.

The comparison of our calculated values of the ratio $\frac{N(H_2^+)}{N(H^+)}$ with the experimental measurements obtained in our laboratory is very satisfactory.

Acknowledgements

References

[1] N.R. Arista, Nucl. Instr. and Meth. B 164–165 (2000) 108.
[2] D.S. Gemmell, Rev. Mod. Phys. 46 (1974) 1.
[3] M.T. Robinson, O.R. Oen, Phys. Rev. 132 (1963) 2385.
[4] J.E. Valdés, P. Vargas, C.D. Denton, N.R. Arista, Nucl. Instr. and Meth. B 193 (2002) 43.
[5] V.A. Khodirev, V.S. Kulikauskas, C. Yang, Nucl. Instr. and Meth. B 195 (2002) 259.
[6] J.C. Poizat, J. Remillieux, J. Phys. B 5 (1972) L94.
[7] N. Cue, M.J. Gaillard, J.C. Poizat, J. Remillieux, J.L. Subtil, Phys. Rev. Lett. 42 (1979) 959.
[8] N. Cue, N.V. de Castro-Faria, M.J. Gaillard, J.C. Poizat, J. Remillieux, D.S. Gemmell, I. Plesser, Phys. Rev. Lett. 45 (1980) 613.
[9] T.R. Fox, Nucl. Instr. and Meth. 179 (1981) 407.
[10] I.S. Bitensky, Nucl. Instr. and Meth. B 103 (1995) 267.
[11] R. Garcia-Molina, M. Barriga-Carrasco, Phys. Rev. A 68 (2003) 054901.
[12] J.E. Valdés, C. Parra, J. Díaz-Valdés, C.D. Denton, C. Agurto, F. Ortega, N.R. Arista, P. Vargas, Phys. Rev. A 68 (2003) 064901.
[13] W. Eckstein, Computer Simulation of Ion–Solid Interaction, Springer-Verlag, Berlin, 1991.
[14] M.J. Puska, R.M. Nieminen, Phys. Rev. B 27 (1983) 6121.
[15] J.C. Ashley, A. Gras-Martí, P.M. Echenique, Phys. Rev. A 34 (1986) 2495.
[16] O.K. Andersen, O. Jepsen, Phys. Rev. Lett. 53 (1984) 2571.
[17] O.K. Andersen, Z. Pawlowska, O. Jepsen, Phys. Rev. B 34 (1986) 5253.
[18] W. Brandt, A. Ratkowski, R.H. Ritchie, Phys. Rev. Lett. 33 (1974) 1325.
[19] J.A. Phillips, Phys. Rev. 97 (1955) 404.
[20] A. Aguado, O. Roncero, C. Tablero, C. Sanz, M. Paniagua, J. Chem. Phys. 112 (2000) 1240.
[21] W. Möller, G. Pospiech, G. Schrieder, Nucl. Instr. and Meth. 130 (1975) 265.
[22] C.D. Denton, I. Abril, M.D. Barriga-Carrasco, R. Garcia-Molina, G. Lantschner, J.C. Eckardt, N.R. Arista, Nucl. Instr. and Meth. B 193 (2002) 198.