

Ion fractions in the scattering of hydrogen on silicon surfaces

Evelina A. García^{*1}, C. González Pascual², P. G. Bolcatto^{3,4}, M. C. G. Passeggi^{1,5}, and E. C. Goldberg^{1,3}

¹ Instituto de Desarrollo Tecnológico para la Industria Química (CONICET-UNL), Güemes 3450, cc91, 3000 Santa Fe, Argentina

² Departamento Física Teórica de la Materia Condensada, C-V, Universidad Autónoma de Madrid, Canto Blanco 28049, Spain

³ Facultad de Ingeniería Química, Universidad Nacional del Litoral, Santa Fe, Argentina

⁴ Facultad de Humanidades y Ciencias, Universidad Nacional del Litoral, Santa Fe, Argentina

⁵ Facultad de Bioquímica y Ciencias Biológicas, Universidad Nacional del Litoral, Santa Fe, Argentina

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We present a theoretical calculation of the resonant charge-exchange process occurring in H^0 scattering by $Si(100)2 \times 1$ surfaces. In the atom-surface interacting system the core states of the surface atoms are included and the parameters of the Hamiltonian are calculated in an *ab initio* basis taking into account the extended features of the surface and the localized atom-atom interactions within a mean-field approximation. The density of states of the surface and sub-surface atoms are obtained from a molecular dynamic-density functional theory in the local density approximation. An elastic binary collision is assumed to fix the projectile trajectory, while the inelastic processes are determined by the interaction of the projectile atom with all the surface atoms 'seen' along its trajectory. The ion fractions are calculated by using the Green-Keldysh formalism to solve the time dependent process. The results, obtained as an average over different possibilities for the scattering center, reproduce the general trends of the experiment.

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1 Introduction

Ion surface collisions have become a useful and reliable tool for studies and analysis of solid surfaces. For many surfaces ion scattering spectrometry (ISS) is capable to discriminate contributions to the backscattering yield coming from both different atomic layers and different atoms present at the surface. A number of experimental and theoretical studies on resonant electron transfer processes have been performed for the case of metal surfaces, although not much has been done concerning with detailed studies on semiconductor structures [1]. A thorough investigation of this process on semiconductors with specific surfaces states playing an important role in the reactivity of these surfaces appears important. Among various possibilities, H^+ is the simplest projectile and the understanding of its interaction with solid surfaces is one of the current topics in surface science. Central to this problem is the dynamics of charge transfer, being the formation of H^- a particular case of resonant electron capture.

Maazouz *et al* [2] have measured the H^- formation in collisions of 1-4 keV beams of H^- and H^+ ions on Si surfaces for a scattering angle of 38° , being the emerging ion fractions investigated in an exit angular range covering from 2° to 36° with reference to the surface plane. As the measurements performed with both H^- and H^+ beams give practically the same negative ion fractions, it is concluded that the charge memory of the incident ion is lost during the incoming trajectory due to an efficient resonant neutralization.

* Corresponding author: e-mail: egarcia@intec.unl.edu.ar

In this work we present results of a theoretical investigation of H^- formation in scattering of H on a Si(100) 2×1 surface. A further work including the Si(111) 7×7 surface is in progress. Realistic local density of states of the reconstructed surfaces were obtained with the Fireball96 code [3]. The energy of the projectile state and hopping terms are obtained from a bond-pair approximation to the projectile-surface interacting Hamiltonian [4]. The Green-Keldysh formalism [5] is used to solve the time dependent evolution of the resonant charge-exchange process.

2 Theory

2.1 The time-dependent Hamiltonian and energy level calculation

By considering the hydrogen affinity level is 3.8 eV above the Si Fermi level (fixed at 0 eV) and that the ionization level can be assumed always occupied since it is resonant with the valence band, one expects that the one-active level (spin-less) approximation should provide a good description of the negative ion formation [6]. In this approximation the Hamiltonian is written as:

$$H = \sum_{K=k,l} \epsilon_K \hat{n}_K + E_\alpha(t) \hat{n}_\alpha + \sum_{K=k,l} [T_{K,\alpha}(t) \hat{c}_K^\dagger \hat{c}_\alpha + h.c.] \quad (1)$$

where the index α refers to the active state of the projectile with energy $E_\alpha(t)$, while the indexes k and l refer to the valence- and core-band states ($1s$, $2s$ and $2p$) of the solid with energies ϵ_K . $T_{K,\alpha}(t)$ represent the corresponding hopping parameters. The time dependence of these parameters comes from the classical trajectory $\vec{R} = \vec{R}(t)$ with constant velocity v . $T_{K,\alpha}$ and short-range contributions to orbital energy, ϵ_α are calculated on an *ab initio* basis by using the bond-pair model [4]. The effect of long-range interactions is introduced by considering for large normal distances (z) to the surface ($z \geq z_0=8$ bohr) an image potential [7], $T_{im}(z) = -(\epsilon - 1)/(\epsilon + 1) \times (1/4z)$. ϵ can be adequately described by the static dielectric constant $\epsilon(0)$ (=11.9 for Si) at low projectiles velocities. As for large velocities the substrate does not have enough time to respond to the field of the moving charge, it is more appropriate to use the effective dielectric constant given by the optical value $\epsilon(\infty)$ (=1.1 for Si) [7]. Besides, it is necessary to contemplate the correct asymptotic behavior of the wave function as seen from the surface reference frame. It means to add the term $v_p^2/2$ in the calculation of the energy level [8] (v_p is the normal velocity to the plane surface). Strictly, this translation factor correction should be also included in the hopping terms. However, it is not expected to have relevant effects at the relative low velocities considered in this work. Consequently velocity effects will be retained only for the atomic energy level position. Under these assumptions $E_\alpha[\vec{R}(t)]$ is then proposed as:

$$E_\alpha(\vec{R}) = \begin{cases} \epsilon_\alpha(\vec{R}) - \epsilon_\alpha(\vec{R}_0) + \epsilon_\infty + T_{im}(z_0) + v_p^2/2 & z < z_0 \\ \epsilon_\infty + T_{im}(z) + v_p^2/2 & z \geq z_0 \end{cases} \quad (2)$$

In this form the energy level obtained within a mean field calculation [$\epsilon_\alpha(\vec{R})$] is shifted to join in a smooth way with the correct asymptotic behavior [ϵ_∞]. This joining procedure has proved to be successful in previous works [9].

2.2 Quantum mechanical calculation of the ion-fraction

In the one-active level spin-less approximation, the average number occupation $\langle \hat{n}_\alpha(t) \rangle$ gives the probability that the projectile state is occupied at time t and $1 - \langle \hat{n}_\alpha(t) \rangle$ gives the probability of the empty state. The time-dependent Green-Keldysh function [5] is used to calculate $\langle \hat{n}_\alpha(t) \rangle$ (see Ref. [9] for details)

$$\langle \hat{n}_\alpha(t') \rangle = \frac{1}{2} [1 - iF_{\alpha\alpha}(t', t')] = \frac{1}{2} [1 + \langle 0 | \hat{c}_\alpha^\dagger(t') \hat{c}_\alpha(t') - \hat{c}_\alpha(t') \hat{c}_\alpha^\dagger(t') | 0 \rangle]. \quad (3)$$

The function $F_{\alpha\alpha}(t, t')$ is calculated by solving the set of coupled equations of motion:

$$i \frac{d}{dt} F_{\alpha\alpha}(t, t') = E_{\alpha}(t) F_{\alpha\alpha}(t, t') + \int_{t_0}^t dt_1 \Sigma^R(t, t_1) F_{\alpha\alpha}(t_1, t') + \int_{t_0}^{t'} dt_1 \Omega(t, t_1) G_{\alpha\alpha}^A(t_1, t'), \quad (4)$$

$$i \frac{d}{dt} G_{\alpha\alpha}^A(t, t') = \delta(t - t') + E_{\alpha}(t) G_{\alpha\alpha}^A(t, t') + \int_t^{t'} dt_1 \Sigma^A(t, t_1) G_{\alpha\alpha}^A(t_1, t'). \quad (5)$$

$G_{\alpha\alpha}^A(t, t')$ being the advanced Green function and the self-energies are given by:

$$\Sigma^A(t, t_1) = [\Sigma^R(t_1, t)]^* = i\Theta(t_1 - t) \sum_{K=k,l} T_{\alpha,K}^*(t) T_{K,\alpha}(t_1) \exp[-i\epsilon_K(t - t_1)], \quad (6)$$

$$\Omega(t, t_1) = -i \sum_{K=k,l} T_{\alpha,K}^*(t) T_{K,\alpha}(t_1) \exp[-i\epsilon_K(t - t_1)](1 - 2n_K). \quad (7)$$

where n_K is the occupation of the solid band states.

3 Results and discussion

The Si(100) 2×1 surface is a reconstruction in which the outermost atoms form dimers. The projectile trajectories correspond to a classical binary collision with each atom of the dimer acting as the scatterer. Thus, we consider two possible trajectories differing in the scattering center. The theoretical ion fractions presented here are an average between these two situations. The collision geometry arrangement for the incoming and outgoing trajectories are roughly simulated by two broken straight lines with the correct incidence and exit angles with reference to the surface plane. The closest distance of approach is determined by using usual concepts of two-body scattering theory [10] from the interaction energy of the projectile-target atom system. Besides, the electronic processes involve the interaction with all surface atoms inside a near-neighbor sphere of radius of 13 bohr centered at the projectile position in each point of the trajectory. In this form the experimental features of scattering geometries are well reproduced.

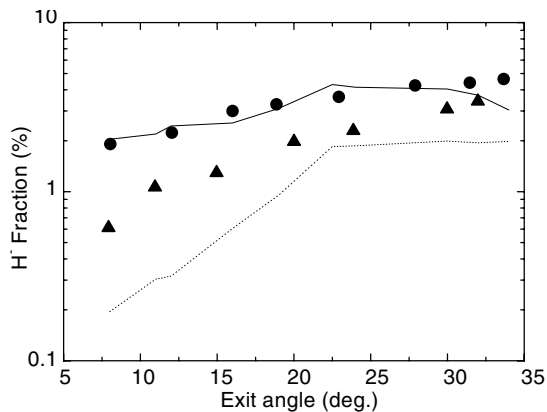


Fig. 1 (a) Ion fractions as a function of the exit angle on the Si(100) 2×1 surface. Points correspond to experimental data from Ref. [2] for incident energy of 4 keV (circles) and 1 keV (triangles). Lines are the theoretical results averaged over the two outer scatterer atoms in the Si(100) 2×1 surface. Solid line for 4 keV and dotted line for 1 keV.

Figure 1 shows comparative results of H^- as a function of the exit angle. $T_{im}(z)$ has been calculated by considering $\varepsilon(0)$ for 1 keV and $\varepsilon(\infty)$ for the 4 keV case. The Si(111) sample used in the experiments is expected to have suffered some amorphization, and the true topology is really unknown. In spite of this fact we observe that our calculations have the same trends of the experimental data at both incident energies.

Figure 2 shows the ion fractions as a function of $v_{p,out}$ (outcoming v_p). As in the experiments, our results are energy-dependent at low velocities, although at intermediate values ($v_{p,out} > 0.05$ a.u.) the capture becomes independent of the kinetic energy of the ion beam. Along the projectile trajectory many surface atoms are contributing in an integrated way to the final charge state of the incoming particle, appearing the charge transfer process more sensitive to the fine details of the surface for small exit angle and for low energy.

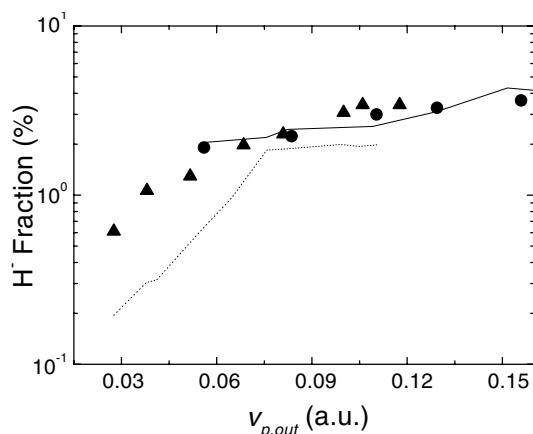


Fig. 2 (a) Ion fractions as a function of the perpendicular velocity. Same conventions as Fig.1

4 Conclusions

In summary, we present a rigorous quantum mechanical calculation for the dynamical H⁻ formation in collisions on Si surfaces. We have considered realistic local density of states, as well as *ab initio* position-dependent parameters including the projectile interaction with much more than first neighbors along the trajectories. Our theoretical results based on a resonant mechanism, where the extended and localized features of the surface electronic structure and the pronounced shift of the projectile level are taken into account, reproduce the general trends of the experiments.

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