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Effect of image potential and charge exchange on the trajectory of fast protons in surface scattering

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Fast hydrogen atoms with an energy of 25 keV are scattered off a clean and atomically flat W(100) surface under grazing angles of incidence. Switching of an electric field directed normal to the scattering plane allows the separation of angular distributions of scattered projectiles leaving the surface as neutral hydrogen atoms and as protons, respectively. We observe a shift for protons towards smaller angles of scattering in direct comparison with neutral atoms. Our experiments give evidence for an effect of image potential and charge exchange on the trajectory of scattered projectiles. The analysis of data allows one to deduce information on charge exchange in atom-surface interactions.

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Scattering of fast atoms or ions from a solid surface is a powerful technique to investigate the vacuum-solid interface [1] and to study charge exchange of atomic species at surfaces [2]. The trajectories for projectiles during the scattering event are determined to a wide extent by effective interaction potentials between projectile and surface atoms. In general, these potentials are approximated by screened Coulomb-type potentials [1,3,4]. In addition, the dielectric response of a conducting surface for charged particles in close vicinity to the surface plane results in an attractive image charge potential. The effect of the image potential in ion surface scattering [5–7] and in ion desorption [8] has been a matter of theoretical treatment. However, no direct experimental observation in this respect has been reported so far.

In this paper we outline experimental studies which given evidence for the effect of the projectile image charge on the trajectory of fast protons in grazing surface scattering. We will show that charge exchange affects the image interaction during collisions with surfaces in a characteristic way. Thus our studies provide information on mechanisms of electron transfer between a surface and atoms.

We will show below that trajectories in surface scattering are most sensitive to image charge effects in grazing incidence geometry. In grazing collisions with surfaces the energy of motion normal to the surface plane (y axis) is given by $E_y = E\Phi_{in}^2$, so that for projectile energies E in the kilo-electron-volt domain and grazing angles of incidence $\Phi_{in} \approx 1°$ E_y amounts to energies typically of electron volts. As a consequence, the projectile will not overcome the collective repulsive interaction potentials with surface atoms and will not penetrate into the bulk of the solid. The projectiles are scattered at the topmost layer of surface atoms, and their trajectories can be approximated by concepts of channeling [9]. From E_y results for the projectile, a distance of closest approach to the topmost layer y_0 which, for our study, amounts to $y_0 \approx 2$ a.u. (atomic units will be used here: 1 a.u.=0.053 nm). Then trajectories can be described by a simple analytical expression [10], and most of the incoming and the outgoing paths can be approximated by a straight line.

In addition to this repulsive interaction potential we have for a charged particle close to a conducting surface an attractive dynamic image potential [5,11], which is described here reasonably well by the classical expression

$$V_{im} = -\frac{1}{4(y - y_{im})}, \tag{1}$$

where y_{im} denotes the position of the effective electronic image plane. From an adjustment of Eq. (1) to the asymptotic limit obtained in nonlocal density-functional calculations, one deduces $y_{im} \approx 4$ a.u. [12].

Since the image interaction is based on the charge of the projectile, it will be affected strongly by charge exchange. The experiments reported here are performed with hydrogen atoms, where the projectiles leave the surface predominantly as neutral hydrogen atoms or as singly charged protons. For protons we expect an effect on their trajectories by the potential given in Eq. (1). However, for neutral atoms the charge of the projectile core is screened to a large extent and contributions of an image interaction can be neglected.

The experimental procedures of our study are connected closely to the concepts of charge transfer in grazing surface collisions [2,13–16]. From the extreme geometry of the collision with the solid results an adiabatic regime of interaction with respect to the motion normal to the surface plane, whereas the high-velocity component of the parallel motion modifies, in the projectile rest frame, the effective population of electronic target states in terms of a "Doppler-Fermi-Dirac distribution" [13]. Charge transfer is dominated by processes of relatively long range: (1) one-electron resonant tunneling and (2) Auger transfer. We will limit our discussion to charge exchange with respect to the $1s$ ground-state term of the hydrogen atom which was found to be populated predominantly after grazing surface scattering [17].

Total transition rates for resonant tunneling [5,18,19] and Auger transfer [19] between a "free-electron metal" and hydrogen $1s$ show an exponential dependence on the distance from the surface. Probabilities for electron capture and loss near the turning point of the trajectory ($y \approx y_0$) are large, and because of the low velocity normal to the surface, charge exchange reaches an equilibrium.

Then a projectile cannot survive in a defined charge state. On the farther path of the outgoing trajectory the projectile reaches a distance y_s ($>y_0$), where the transition rates are still sufficiently high for electron capture, however, already too low for subsequent electron loss. It turns out that the final formation of atomic terms takes place within a rather defined interval of distances around the so-called "freezing distance" y_s, where the final charge state is formed ("frozen out") [20]. In a recent theoretical investigation of proton neutralization into hydrogen 1s during grazing surface collisions $y_s \approx 7$ a.u. is reported [15].

At distances around y_s the projectiles reach their final charge states, capture or loss events on the remaining part of the trajectory ($y > y_s$) are unlikely. As a consequence, a defined image interaction can take place only at distances $y > y_s$. Then the energy of motion along y for a proton is given by

$$E_y = E\Phi_{\text{out}}^2 + V_{\text{im}}(y_s) , \qquad (2)$$

whereas for neutral atoms the image term vanishes. The modification of E_y due to image interaction results in an angular shift $\Delta\Phi_{\text{out}}$ towards the surface plane for trajectories of projectiles emerging as protons. In comparison with neutral atoms scattered from the surface under Φ_{out} the distribution of protons is shifted by

$$\Delta\Phi_{\text{out}} = \Phi_{\text{out}}\{1 - [1 + V_{\text{im}}(y_s)/E_y]^{1/2}\} . \qquad (3)$$

For $E_y \gg V_{\text{im}}(y_s)$ Eq. (3) can be approximated by $\Delta\Phi_{\text{out}} \approx V_{\text{im}}(y_s)/2E\Phi_{\text{out}}$, which shows that a grazing scattering geometry is favorable to detect image potential effects.

The experimental procedure used here is the separation and the detection of angular distributions for projectiles scattered from the surface as protons and neutral atoms. Image charge effects manifest themselves by an angular shift between the two distributions. Basic features of the experimental setup are sketched in Fig. 1. A beam of 25-keV hydrogen atoms is collimated by a set of slits to a divergence of about 0.02° and directed onto the (100) face of a tungsten monocrystalline target under grazing angles of incidence $0.2° \leq \Phi_{\text{in}} \leq 1.5°$. In order to avoid effects of an axial surface channeling, a high-index crystallographic direction in the surface plane of the target is oriented along the beam axis ("random" direction). The experiments are run under UHV conditions at a base pressure of some 10^{-11} mbar. The target surface is carefully polished to match the (100) plane as close as possible and prepared *in situ* by cycles of grazing sputtering with 25-keV argon ions at $\Phi_{\text{in}} \approx 2°$ and annealing or flashing up to temperatures of about 1800 °C. Cleanness of the surface is inspected by Auger spectroscopy. Analysis of low-energy electron-diffraction (LEED) spot profiles by a spot profile analysis (SPA) LEED system [21] yields an average terrace width formed by surface atoms > 80 nm at an average step height of half a lattice constant (0.16 nm). Such a high-quality surface is crucial for the studies reported here.

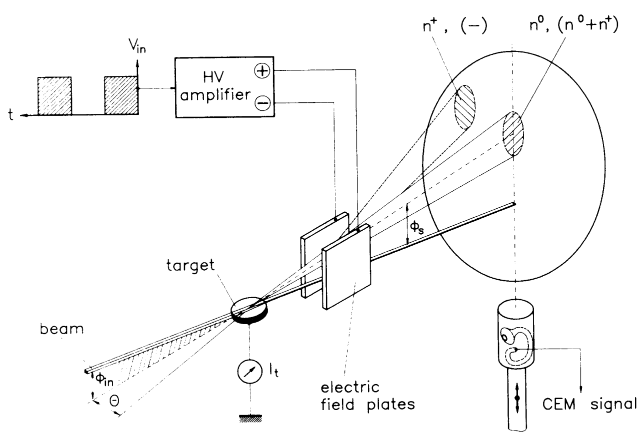

FIG. 1. Sketch of the experimental setup to separate the angular distributions of projectiles scattered in two different charge states: n^0 is the signal due to neutral atoms and n^+ the signal due to protons. The two distributions are isolated by a "difference method" (see text). HV denotes high voltage and CEM channel electron multiplier.

Polar distributions of scattered projectiles are obtained with a channeltron detector which is positioned 700 mm behind the target by means of a precision manipulator with a stepmotor drive. A diaphragm of 0.4 mm diameter in front of the channeltron covered with a thin carbon foil (5 μg/cm^2) provides a good angular resolution and a constant response of the detection to different charge states. The separation of the angular distributions for neutral atoms and protons is performed with the help of an electric field of about 500 V/cm directed normal to the polar plane of scattering. In order to avoid a modification of the shape of the distributions for protons due to slight imperfections and higher-order moments of the electric field, we apply a *difference method* to obtain both distributions with high reliability.

We record first the signal without electric field and obtain contributions from all charge states ($n^0 + n^+ + n^-$); the yield of negative ions n^- is of the order of 10^{-3} and will be neglected henceforth. Afterwards a voltage is applied to the field plates, so that the signal at otherwise unchanged conditions stems only from neutral atoms (n^0). So the difference between both signals allows the isolation of the contributions of protons (n^+). In order to keep effects due to fluctuations of the projectile beam, etc., on the data small, we switch the two settings of the electric field at a frequency of several hertz and register the corresponding signals in separate channels. By this technique we record complete polar distributions of scattered projectiles via microcomputer control.

A typical result is shown in Fig. 2. On the left side we display the dependence of the signal on the angle of scattering Φ_s for a negligible voltage on the field plates; on the right side the corresponding signal, when a voltage $U = -840$ V is applied to one of the plates. In both distributions we find a strongly saturated peak on the left margin caused by a fraction of the residual beam, which passes over the target without interaction (see also Fig. 1). This peak serves as a reference for the direction of the projectile beam. The other peak results from projectiles which are scattered from the target surface. In the final state of preparation of the target, we observe half-widths of the angular distributions down to about 0.5°, i.e., the

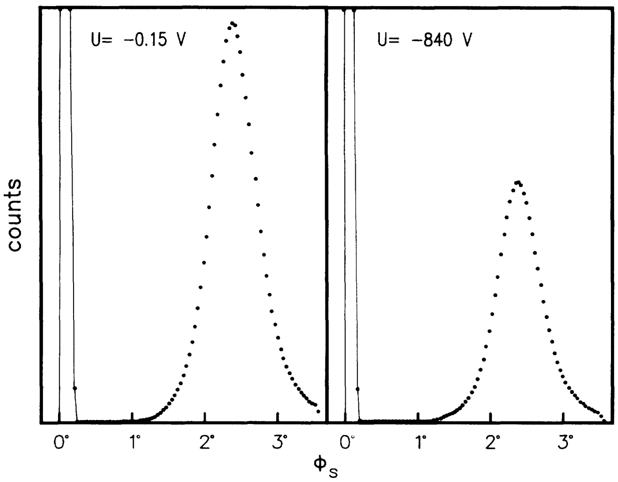

FIG. 2. Signal of the channeltron detector dependent on the angle of scattering Φ_s obtained after the interaction of a 25-keV beam of hydrogen atoms with a clean W(100) surface. The left side of the figure shows data for a voltage $U=-0.15$ V applied to the electric-field plates; the right side shows data for $U=-840$ V. The saturated peak on the left sides of both curves is due to a fraction of the projectile beam that has passed over the target without deflection and indicates the direction of the projectile beam. Note that this peak is not affected by the different settings of the electric field which demonstrates that the projectile beam is neutral.

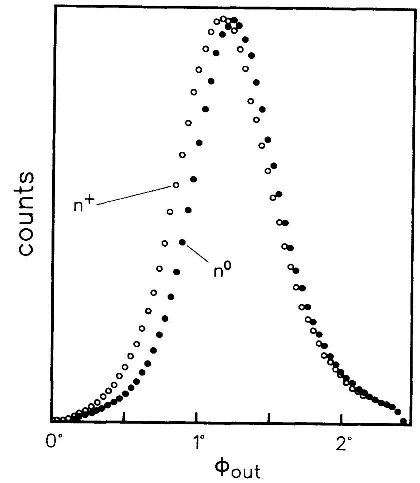

FIG. 3. Dependence of the signals due to protons (open circles) and neutral hydrogen atoms (solid circles) on the angle of emergence Φ_{out}. The data for the protons (n^+) are obtained by taking the difference from the two data sets shown in Fig. 2. The distributions are normalized in the plot to the same maximum.

projectiles are scattered within narrow geometrical limits. Investigations with SPALEED indicate that the half-width is closely related to the defect structure of the target surface. The densities of defects and also the half-widths are found to increase with the dose of irradiation by the projectile beam, so that the sample has to be annealed by flashing to temperatures up to about 1800 °C for 45 s after periods of about 10 min of irradiation with the projectile beam.

In Fig. 3 we present a direct comparison between the polar distributions for neutral atoms (solid circles) and for protons (open circles). The data for protons are obtained by taking the difference between the two data sets shown in Fig. 2. Both distributions are normalized to have the same maximum and are plotted versus the angle of emergence $\Phi_{out}=\Phi_s-\Phi_{in}$. It is evident from the figure that the distribution for protons is shifted towards smaller angles Φ_{out} and is broadened slightly in comparison to the data for atoms. The maxima of the two distributions are separated by $\Delta\Phi_{out}\approx 0.1°$. Our results demonstrate that the experimental procedures applied here allow one to resolve such a small shift in polar angles.

With the very same settings of collimation and target we scatter also a beam of a He-Ne laser from the surface in order to obtain the direction of specular reflection. Then we find for a neutral projectile beam (no image interaction on the incoming path) that the maximum of the distribution for projectiles emerging from the surface as neutral atoms coincides with the maximum of the reflected laser beam intensity. Thus the maximum of n^0 in Fig. 3 at $\Phi_{out}=1.3°$ represents the specularly reflection portion of projectiles. From the angular shift $\Delta\Phi_{out}$ when these projectiles emerge from the solid as protons, we obtain from Eq. (3) $V_{im}(y_s)\approx -0.07$ a.u. This corresponds to an effective freezing distance $y_s\approx 7.5$ a.u., according to Eq. (1).

At the present level of evaluation of data the good agreement with the theoretical estimate should not be overestimated. However, the consistency with theoretical concepts of charge exchange may serve as an indication that our experimental technique provides new information on details of electronic transfer between atoms and solids. In this respect we note that the somewhat larger angular width of the distribution for protons can be ascribed to the finite interval of final charge exchange around y_s. A more refined analysis of the data is in progress [22] and allows one to reproduce the broadened distribution for protons by concepts of the "freezing approximation" [2,20,23]. In this approximation y_s decreases slightly with the increasing normal velocity component, i.e., with the increasing angle of emergence Φ_{out} $V_{im}(y_s)$ is expected to increase also. Our experimental data seem to show a corresponding increase within the range of Φ_{out} studied here.

In conclusion, in grazing scattering of fast hydrogen atoms from W(100) we observe an angular shift between polar scattering distributions for projectiles emerging in different charge states. These data give experimental evidence of an effect of the image charge on the trajectory of projectiles in surface scattering. We find that the image interaction is affected in a characteristic way by charge-exchange phenomena in the vicinity of the surface plane. The data can be interpreted by current theoretical concepts of electron transfer in grazing surface collisions and allow one to deduce effective distances of the final formation of atomic terms. We hope that this work will stimulate more refined theoretical treatments that will take into account the perturbation of the effective trajectory of the projectile by electron-transfer processes during the complete scattering event.

Our observation of image charge effects in grazing incidence geometry also allows one to extrapolate that contributions of these effects for ion scattering spectroscopy are generally small. The demonstration of an effect of the attractive image force on the trajectory of fast ions gives support to the model of "skipping motion," a quasibinding of ions by image interaction to the surface plane, proposed by Ohtsuki and co-workers [5,6,24]. Experimental evidence for those hopping types of trajectories at the surface has been reported recently by discrete losses in the energy spectra of protons scattered from a graphite surface [25]. Similar results are obtained by Winter and Sommer for the interaction of fast protons and hydrogen atoms with an Al(111) surface [26].

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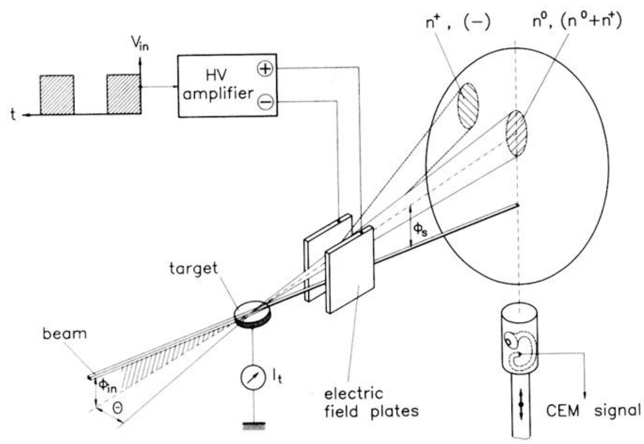

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