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Neutralization of fast protons in grazing collisions with a clean Al(111) surface

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We investigate the neutralization of protons scattered from a clean Al(111) surface under grazing incidence for projectile energies ranging from 50 keV to 1.25 MeV. The neutral fraction of the scattered beam strongly depends on the state of preparation of the target surface and monotonically decreases with increasing projectile energy. The fractions are generally smaller than observed after transmission through thin aluminum foils. Our experimental data do not agree with recent theoretical predictions claiming dominant contributions of second-order Thomas scattering to neutralization.

Collisions of fast ions with surfaces under grazing incidence provide good conditions for well-defined studies of charge-exchange mechanisms between atoms and surfaces. Experiments in this geometry are run under semiplanar channeling conditions; i.e., the interaction of projectiles with the solid is characterized by two vastly different time (velocity) regimes: the motion parallel to the surface plane with projectile energy E (velocity v) and normal to the surface with energy $E\sin^2\Phi$ ($v\sin\Phi$). For typical grazing angles of incidence used in this study ($\Phi=3$ mrad) the energies of normal and parallel motion differ by about 5 orders of magnitude. As a consequence even at the largest energies of our experiments (~ 1 MeV) the projectiles are predominantly scattered at the surface plane in a large number of small-angle scattering events without penetration into the bulk of the solid, giving rise to well-defined trajectories during charge transfer with the solid.

The charge-exchange processes at the surface are directly affected by the type of trajectory: in the present case there is a quasiadiabatic regime with respect to the perpendicular motion and, in contrast, a regime of fast ion-surface collisions with respect to the parallel motion. Since the projectiles escape from the surface at low normal velocities, mechanisms of charge exchange with a relatively long range are expected to dominate the final formation of atomic terms, i.e., one-electron resonant tunneling and the Auger process. Both mechanisms lead to neutralization as well as ionization of atoms near surfaces, where the contributions to electron capture and loss depend on the binding energies of atomic terms, the work function of the surface, the Fermi energy, and the distribution of occupied and unoccupied electronic states of the conduction band.

In a number of recent papers it has been shown that for projectile velocities $v < v_0$ (Bohr velocity) concepts of resonant tunneling result in a good description of H^- formation,[1,2] neutralization of alkali ions,[3] and anisotropic formation of excited atomic terms.[4,5] For atoms with binding energies clearly larger than the work function of the solid (e.g., the hydrogen $1s$ term) contributions to charge exchange via Auger neutralization as well as Auger ionization become important.[6]

In electron transfer in grazing surface collisions the effects due to the high parallel velocity play an important role, because in the rest frame of the atom the density of occupied and unoccupied conduction-band states is modified by this motion in terms of a "Doppler-Fermi-Dirac distribution".[1-9] This modification of the effective density of metal states affects charge transfer due to electron capture and loss in a characteristic way.[3,5,7,9] As one consequence one finds, for velocities larger than v_0, that resonant neutralization and to a lesser extent also Auger neutralization play practically no role for electron capture. However, ionization by both processes is still effective because of the available phase space of unfilled metal states.

In analogy to ion-atom collisions, neutralization may then proceed via capture in broadened resonances from localized inner-shell levels of the target and to some extent via higher-order processes of charge transfer. Thumm and Briggs[10] have recently presented an analysis of such higher-order processes in electron capture from a surface by grazing incidence impact of protons with velocities larger than v_F (Fermi velocity). Their calculations imply that the second-order Thomas mechanism, i.e., a double scattering process of target electrons with projectile core and target core,[11] exceeds the results of calculations in first Born approximation[12] by about 2 orders of magnitude. Since Thomas-scattered electrons have typical energies used in low-energy electron diffraction (LEED), structures in the monotonic decrease of the neutral fraction with projectile velocity are expected. Our paper describes the first experimental tests of the theoretical predictions given in Ref. 10.

The experiments are performed with protons of energies ranging from 50 keV to 1.25 MeV at the 2.5-MV van de Graaff accelerator of the Institute de Physique Nucleaire, Lyon (France). The projectile beam is collimated by sets of diaphragms (width ~ 0.2 mm) to a sub-mrad divergence and is then scattered from a clean Al(111) surface under grazing angles of incidence of about 3 mrad.

Differential pumping on both ends of the target chamber results in a base pressure in the upper 10^{-8} Pa domain during the runs. The (111) face of an aluminum monocrystalline sample was polished with great care by keeping the deviation between the (111) plane and polishing plane as small as possible (<2 mrad) to achieve a low density of steps at the surface, i.e., an average width of terraces formed by surface atoms of better than about 75 nm. Impurities at the surface (especially oxygen and carbon) are removed by grazing sputtering ($\Phi_{\text{in}} \approx 1°$) with 400-keV Ar^+ ions of about 1–2 $\mu\text{A/mm}^2$ current density. Preparation of the target by frequent cycles of sputtering and annealing by heating the crystal up to 560 °C finally yields a clean and flat surface. After such a treatment no indications stemming from impurities can be found in the Auger spectra. We will demonstrate below that the "quality" of the target surface drastically affects the neutral fraction of the scattered beam.

During sputtering the target is rotated around its surface normal within 360° under simultaneous recording of the uncompensated target current. This current is mainly due to kinetic emission of electrons, and is observed to increase slightly if the beam is axially channeled in a low-index direction at the crystal surface.[13] By this simple on-line technique for positioning the target with respect to an azimuthal orientation, it was checked that the projectile beam is directed along a high-index axis ("random" direction) to avoid axial surface channeling effects.

In Fig. 1 we display a distribution for 525-keV protons scattered in a polar plane which is obtained with the help of a detector (0.2-mm diaphragm mounted in front of a channeltron) 1 m behind the target. The saturated peak on the far left-hand side stems from the residual beam that has passed above the target without scattering and represents the direction of the projectile beam. The distribution of scattered projectiles is well defined, a half-width of typically 4 mrad being observed. The separation between the two peaks in Fig. 1 defines the angle of scattering for the specularly reflected part of the beam: $\Phi_s \approx 6$ mrad. We also have shown in Fig. 1 the neutral fractions n^0 for different angles of scattering (dots with error bars). These fractions are obtained by selecting corresponding angles with the help of a slit of 0.2 mm width positioned by a linear feedthrough 150 mm behind the target (resolution about 1 mrad), and consecutive separation of charge states by means of a pair of electric-field plates. Since only a very small fraction of the beam gets neutralized at the energies used in this study, great care has to be devoted to saturation effects of the detector which enhance the ratios of low to high count rates. We deduce from data, as shown in Fig. 1, that the neutral fractions have a slow dependence on the scattering angle. Consequently, we will refer to specularly reflected projectiles with respect to all data given below.

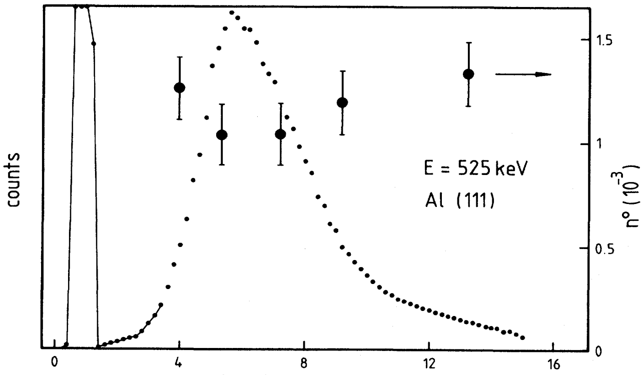

FIG. 1. Angular distribution of 525-keV protons scattered at an Al(111) surface. The dots with error bars represent neutral charge fractions.

A striking feature found in our experiments is a pronounced sensitivity of the neutral fractions on the state of preparation of the target. In Fig. 2 we show at the right-hand side a typical result obtained for 825-keV protons scattered at a target in the final state of preparation (sputtering and annealing with temperatures between 450 and 500 °C). This fraction is strongly enhanced when the same experiment is repeated after a time which allows it to build up some coverage of the surface with adsorbates. That case is usually met in a continuation of previous runs on the following day (see left-hand side of Fig. 2). The neutral fractions are even further enhanced by subsequent annealing of the samples, which also implies that the neutralization is enhanced by the presence of adsorbates at the surfaces. After removal of this coverage by sputtering and after cycles of sputtering and heating, overall consistent data are obtained in different runs (right-hand side of figure).

The dependence of the neutral fractions on projectile energy and velocity is displayed in Fig. 3. The fractions are generally small and monotonically decrease by about 3 orders of magnitude within the range of velocities investigated here ($1.4v_0$–$7v_0$). For comparison we also show data obtained after transmission of protons through thin

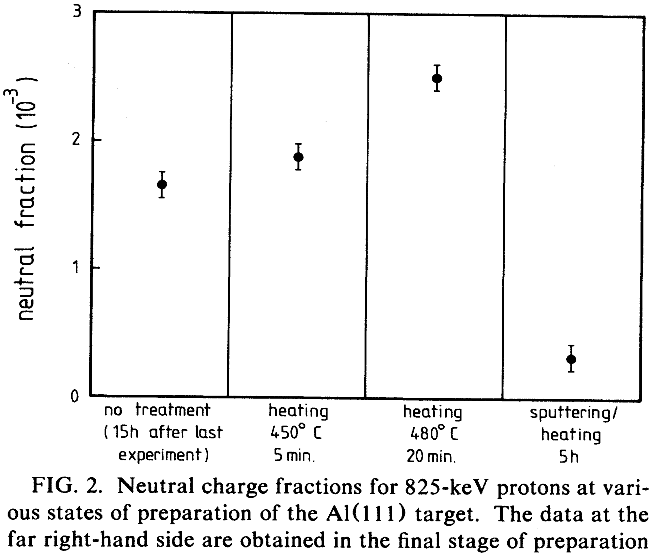

FIG. 2. Neutral charge fractions for 825-keV protons at various states of preparation of the Al(111) target. The data at the far right-hand side are obtained in the final stage of preparation of the target by cycles of sputtering and annealing with temperatures between 450 and 500 °C. At the left-hand side we show typical data after the experiment has been stopped for 15 h and after two different subsequent periods of heating of the target.

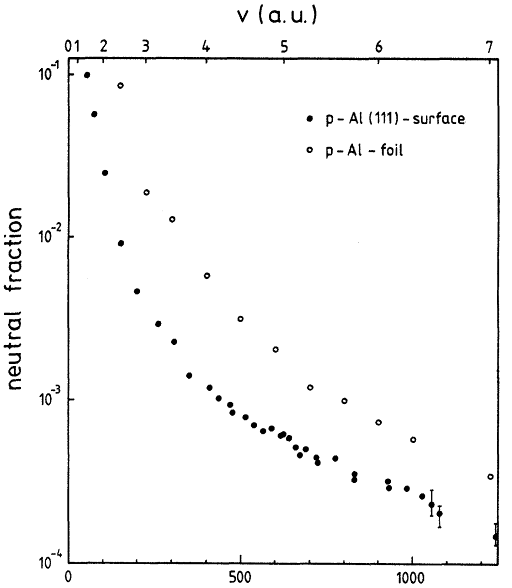

FIG. 3. Neutral charge fractions for specularly reflected protons ($\Phi_{\text{in}} \approx \Phi_{\text{out}} \approx 0.2°$) in dependence on projectile energy/velocity. Typical uncertainties in the data are indicated at the high-energy end. The open circles represent beam-foil data as obtained by Ref. 13.

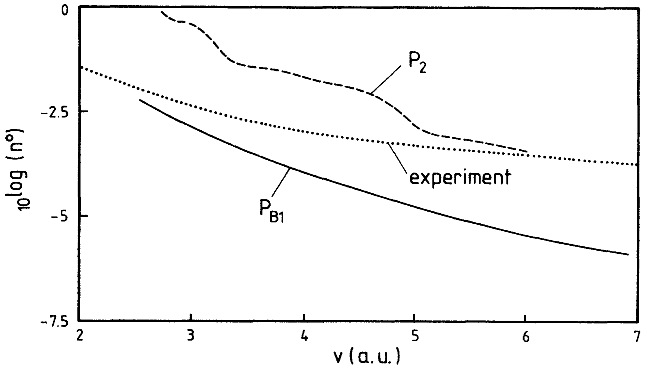

FIG. 4. Comparison of our experimental results (represented by the dotted curve) with theory of Ref. 10. The solid curve represents first-order contributions; the dashed curve represents second-order Thomas scattering.

aluminum foils from Ref. 14 (open circles). It is evident from Fig. 3 that the neutral fractions observed after grazing surface scattering are factors of about 3–5 smaller than after beam foil. However, both data sets run about "parallel" and show a comparable dependence on energy. It is interesting to note that neutral fractions after channeling through thin crystals are also found to be smaller than after random beam-foil interaction.[15] The reduction, however, is not as pronounced as for surface scattering. Since the interaction times with the target surface are much larger in grazing scattering, the lower neutral fractions in comparison to beam foil may be surprising at first glance. However, this feature can be simply understood by pronounced ionization because of the energetic resonance between the atomic term and unoccupied metal states.

In Fig. 4 we compare our data (represented by the dotted curve) with recent calculated probabilities of capture by Thumm and Briggs.[10] The solid and dashed curves show theoretical results for neutralization via first- and second-order processes, respectively, in the interval of velocities from $3v_0$ to $7v_0$. According to the calculations, second-order Thomas scattering dominates electron capture and yields steplike structures in the velocity dependence due to LEED-type processes. However, aside from a poor agreement with our data on an absolute scale, no steplike structures at all are observed.

In conclusion, our experiments provide data with respect to neutralization of fast protons via grazing surface scattering under well-defined conditions and will challenge further theoretical efforts on this subject. In this respect future model calculations have to investigate contributions of capture in broadened resonances from localized inner-shell levels of the target[5] as well as electron-loss processes with relatively long range that reduce the effects of capture processes in close collisions with surface atoms.

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[1] J. N. M. van Wunnik and J. Los, Phys. Scr. **T6**, 27 (1983).

[2] J. J. C. Geerlings, J. Los, J. P. Gauyacq, and N. M. Temme, Surf. Sci. **172**, 257 (1986).

[3] R. Zimny, H. Nienhaus, and H. Winter, Radiat. Eff. Defects Solids **109**, 9 (1989).

[4] R. Brako, Phys. Rev. B **30**, 5629 (1984); R. Brako, H. Winter, and D. M. Newns, Europhys. Lett. **7**, 213 (1988).

[5] J. Burgdörfer, E. Kupfer, and H. Gabriel, Phys. Rev. A **35**, 4963 (1987).

[6] H. Nienhaus, R. Zimny, and H. Winter, Radiat. Eff. Defects Solids **109**, 1 (1989); R. Zimny and Z. Miscovic (unpublished).

[7] D. M. Newns, Comments Condens. Matter Phys. **14**, 295 (1989).

[8] H. Schröder, Nucl. Instrum. Methods B **2**, 213 (1984).

[9] H. Winter and R. Zimny, in *Coherence in atomic collision physics*, edited by H. J. Beyer *et al.* (Plenum, New York, 1988), p. 283.

[10] U. Thumm and J. Briggs, Nucl. Instrum. Methods B **43**, 471 (1989); U. Thumm, Ph.D. thesis, Freiburg, 1989 (unpublished).

[11] L. H. Thomas, Proc. R. Soc. London A **114**, 561 (1927).

[12] D. Belkic and R. Gayet, J. Phys. B **10**, 1911 (1977).

[13] H. Winter, Ph.D. thesis, München, 1986 (unpublished).

[14] A. Chateau-Thierry and A. Gladieux, in *Atomic Collisions in Solids*, edited by S. Datz *et al.* (Plenum, New York, 1975), p. 307.

[15] M. J. Gaillard, J. C. Poizat, J. Remillieux, A. Chateau-Thierry, A. Gladieux, and W. Brandt, Nucl. Instrum. Methods **132**, 547 (1976).