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Systematic analysis of different experimental approaches to measure electronic stopping of very slow hydrogen ions

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ABSTRACT

The electronic stopping cross section (SCS) of Ni for slow H^+, H_2^+, D^+ and D_2^+ ions has been investigated for different types of targets in two complementary experimental geometries, i.e., in transmission and backscattering. To warrant sample purity, both a high purity nickel sheet and nanometer Ni layers were prepared in-situ under ultra-high-vacuum conditions. In an alternative approach, ultra-thin samples were prepared ex-situ as self-supporting foils and as nanometer films on a polished substrate (silicon). Identical SCS results are obtained in backscattering using the in-situ prepared film and the high purity sheet. The ex-situ prepared targets contained considerable concentrations of impurities of low atomic numbers, whose contribution to the SCS can be rectified by applying Bragg's rule using TRIM stopping for the impurities. In this way for the ex-situ targets the accuracy of the resulting SCS data is improved considerably. Concordant stopping cross section data are obtained in both geometries. The achieved accuracy does, however, not permit to spot a possible influence of different impact parameter regimes explored in transmission and in backscattering geometries.

1. Introduction

Ions propagating in solids are slowed down due to interaction with both, nuclei and electrons, i.e. by nuclear and electronic stopping, respectively. The mean energy loss per path length is given by the stopping power $S = dE/dx$. In other words, S is the deceleration force acting on the ion. When interactions of ions with atoms or molecules are investigated, the stopping cross section $\varepsilon = S/N$ is a useful quantity, where n stands for the atomic or molecular density of the target material. Profound understanding of the underlying physical processes as well as accurate experimental data are required in many different fields, e.g., space weathering [1], nuclear fusion research [2], and materials research (ion implantation, ion beam analysis) [3,4].

At high ion velocities $v \gg v_F$ (v_F denotes the Fermi velocity of the target electrons), energy dissipation of the projectile is mainly due to electronic stopping and accurate theoretical models are available [5–9]. At low ion velocities $v < v_F$, electronic stopping is due to interaction with the valence electrons; also nuclear stopping may contribute considerably to the total energy loss. The presence of the slow ion in the target material represents a strong perturbation in the states of valence and conduction electrons. Consequently, any theoretical description of electronic stopping of slow ions constitutes a complex many-body problem. Hence, the physics of electronic interactions of slow ions with solids is still a subject of current research [10–12].

Also from an experimental point of view it is demanding to obtain accurate stopping data for slow ions: in recent years, SCS acquired by complementary experimental techniques (transmission and backscattering) exhibited rather pronounced differences in some cases (e.g., Cu and Ag, [13–15]) while for others concordant results were reported (e.g., Al and Au, [16,17,13,18]). The observed differences might point towards a systematic influence of the experimental geometry, due to different impact parameter selection in transmission and in backscattering. This point has recently been discussed thoroughly by Sigmund and Schinner [19].

In order to scrutinize what are the possible reasons for the observed differences between low-energy transmission and backscattering experiments, a cooperation has been started between the Centro Atómico Bariloche and the Johannes-Kepler-Universität Linz (JKU). In a similar

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cooperation, Mertens et al. showed that around the stopping maximum consistent stopping data could be obtained by transmission and by backscattering measurements, when the influence of surface contaminations was properly taken into account for the foils used in transmission measurements [20]. In this study, the electronic stopping of slow H ions in Ni was chosen as benchmark test. As a transition metal, Ni is interesting from a physical point of view, but it is also chemically reactive and therefore provides information on the relevance of target impurities of low atomic numbers (low Z impurities, e.g., carbon, oxygen).

With the aim of achieving accurate electronic stopping cross section data, systematic errors had to be minimized in target preparation as well as in energy loss measurements, both in transmission and in backscattering geometry.

The paper is structured as follows: in Section 2, theoretical models to describe electronic stopping of slow ions are discussed briefly, followed by Section 3, in which simulation programs used in data evaluation of transmission and backscattering experiments are presented. In Section 4, both experimental approaches as well as the techniques used for sample production and characterization are explained in detail. The experimental results are presented and discussed in Section 5.

2. Theory

Since at low ion energies, electronic stopping is due to the excitation of conduction/valence electrons, Ni ($[Ar]3d^8 4s^2$) exhibits an interesting electronic density of states (DOS), with a high DOS at energies below the Fermi level, E_F [21]. In this velocity regime, it has been shown that for a Free Electron Gas (FEG) the stopping power is proportional to the ion velocity, $dE/dx = Q \cdot v$ [22]. The friction coefficient, Q, of an ion of atomic number Z_1 in a FEG has been determined in a nonlinear model within the Density Functional Theory (DFT) framework [23,24], resulting in $Q = n_e \cdot m_e \cdot v_F \cdot \sigma_{tr}$, with the electronic density n_e, and the transport cross section σ_{tr}. This description considers the conduction electrons of a metal as a FEG with a density-related r_s parameter, $r_s = (3/4\pi n_e)^{1/3}$; the Fermi velocity is given by $v_F = 1.919/r_s$. It has been shown empirically that the nonlinear DFT model [23] successfully describes experimental stopping data for protons at $v \leq v_F$ for a wide range of materials, when effective r_s values (deduced from plasmon energies) are used [25]. For the present work we have employed the DFT results for $Q(r_s)$ from Ref. [26].

3. Simulations

The large scattering cross sections at low ion energies lead to an increasing probability for multiple scattering and, consequently, to an increase of nuclear stopping and the path length inside the target, respectively [27]. Therefore, evaluation of data from both, transmission and backscattering experiments needs to rely on Monte Carlo (MC) simulations which allow for an adequate consideration of multiple scattering and permit to disentangle contributions from nuclear and electronic stopping, respectively.

Simulations of energy loss spectra acquired in transmission were performed using a MC code, which was employed already in previous investigations [28–30]. These simulations include information on surface roughness of the target foil, stopping forces, intrinsic straggling and a screened scattering potential of Molière type [31], with the aim of analyzing and explaining the main features of the experimental data. The screening length is determined from the analysis of multiple-scattering angular distributions, and the electronic stopping force results from the comparison of the simulations to experimental angular dependent energy loss spectra [28,32].

For the evaluation of backscattering experiments, MC simulations of energy spectra were performed employing the TRBS code [33], which permits to simulate multi-layer targets. In the simulations, for the individual layers electronic stopping (from TRIM85 [34]) and the strength of the scattering potential (ZBL [34] or Molière [31] screening) can be optimized, e.g. following Ref. [35]. A proper choice of these parameters is essential for accurate evaluation of electronic stopping from energy spectra of backscattered ions [36]. When the stopping power is evaluated by comparing the backscattering spectra of the material of interest and of a reference material of similar atomic number, the scattering potential does not introduce any noticeable systematic errors [37]. In this contribution, the ZBL potential without a screening length correction was applied. Details concerning the evaluation procedure are explained in the experimental section.

4. Experiment

4.1. Target preparation and characterization

4.1.1. Transmission foils

The self-supported ultra-thin foils used in the transmission experiments were produced ex-situ by sputter deposition of Ni films on cleaved polished NaCl crystals under high vacuum conditions. After careful dissolution of the salt by deionized water at 49 °C, the foils floating on the surface were transferred to TEM grids. The thickness and the composition of the films were determined by use of twin samples deposited onto SiO_2 and glass, thereby disregarding possible influences of the preparation of the free-standing foils (incorporation of impurities, oxidation).

The geometrical thickness of the films was measured by low-angle X-ray reflectometry (XRR), which evaluates the interference pattern to yield the geometrical thickness of the Ni film; surface contaminations contribute only little due to the lower sensitivity of XRR to low Z surface impurities. This technique is little sensitive to impurities present in the bulk of the films. Therefore, a mean geometrical film thickness is obtained, but the film density due to incorporated impurities has to be determined independently. The investigated surface area is $\sim 1\ cm^2$ and yields information on film roughness. Possible systematic errors may result from not perfectly plane substrates, too large film roughness, bulk impurities or misalignment. The statistical uncertainty arises from the evaluation of peak positions and depends on the number of visible diffraction orders. Independently, the roughness (including surface contamination) has been checked by Atomic Force Microscopy (AFM). Calibration of the AFM by scanning an MCNC step-height reference of 44 nm under the same experimental conditions as in the present measurements permitted to evaluate the geometrical thicknesses of the films, with an estimated precision of ~5 to 10% (standard deviation).

Depth profiling by means of X-ray photoelectron spectroscopy (XPS) during Ar bombardment yielded information on surface and bulk contaminations, as well as information on the chemical environment of the Ni atoms; the results are presented in Section 5.

4.1.2. Backscattering films

For the backscattering experiments three types of Ni samples were used. First, a set of ultra-thin Ni films was deposited ex-situ by e-beam evaporation on Si under high vacuum conditions. Second, ultra-thin Ni films were deposited in-situ on B/Si using an Omicron EFM-3T evaporation system in ultra-high vacuum (UHV). Note that during evaporation the pressure in the vacuum chamber never exceeded $\sim 2 \times 10^{-10}$ mbar (accomplished by use of a 99.99% Ni rod as evaporation material and two LN_2 cold traps, respectively). Third, a high purity Ni sheet[1] was mechanically polished, cleaned in isopropanol in an ultrasonic bath, and sputter-cleaned by 3 keV Ar^+ ions in the UHV setup. Purity of the in-situ Ni films, the Ni sheet and a reference sample (Cu) was checked by Auger Electron Spectroscopy (AES), the ex-situ Ni films were analyzed by XPS at ZONA (JKU); the XPS results are presented in Section 5.

[1] According to time-of-flight Elastic Recoil Detection (TOF-ERD), performed at Uppsala University, the amount of all bulk impurities was < 1%.