

Electronic interactions of medium-energy ions with solids: some recent results and their implications for high-resolution depth profiling

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Both qualitative and quantitative understanding of the electronic interactions of ions at energies of a few up to several hundred keV is of decisive importance for quantitative material analysis employing ions in the present energy regimes. For example, only accurate knowledge on charge exchange processes permits to employ a method like Low-energy Ion Scattering as a tool to probe the composition of the outermost atomic layer of solids. On a similar basis, inaccuracies in the description of the energy loss experienced along ion trajectories in the solid lead to difficulties in establishing quantitative depth profiles in e.g. Medium-Energy Ion Scattering (MEIS) or other near-surface depth profiling techniques.

Although investigated for a long period of time an accurate prediction of the absolute magnitude of the energy loss per unit path length, the stopping power $S = dE/dx$, is often not straightforward possible. This complication has several reasons: at first, the increased contribution from nuclear losses requires more advanced evaluation routines for deducing reference data, as compared to the MeV-energy regime. Second, the description of the electronic energy loss is complicated by the increased weight of the valence electrons which are sensitive to the chemical surrounding and by the fact, that the interaction is no longer adiabatic, requiring consideration of e.g. dynamic screening phenomena or complex charge exchange processes. Finally, for thinner targets higher relative weight of the near-surface regions of the target materials makes high standards during preparation and accurate knowledge on sample composition an ultimate requirement for obtaining high quality data.

In this contribution recent studies of the energy loss of light and heavy ion in the energy regime from about 10 to about 300 keV performed at Uppsala University are presented to exemplify some of the above mentioned complications in analysis, the complexity of the obtained results as well as the necessity of extensive sample characterization, also for relatively simple systems.

As a first example, a study of the energy loss of Ne ions in Au, Pt and Ag is presented [1]. The presented investigations were performed to obtain a better understanding of the relative contributions of electronic and nuclear energy losses to the experimental spectra. Thin films of the materials of interest were prepared by sputter deposition on Si-substrates. Thickness calibration and characterization of the thin films was performed ex-situ by means of Rutherford Backscattering Spectrometry (RBS) and Time-of-Flight Elastic Recoil Detection Analysis (ToF-ERDA). Ne ions with energies between 80 and 280 keV were scattered from the targets and detected in backscattering direction by a position-sensitive detector with a large solid angle in a Time-of-Flight Medium Energy Ion Scattering (ToF-MEIS) set-up [2]. The present approach allows for high energy resolution and only minor sample deterioration due to the low employed primary particle dose. Spectra of backscattered particles were analyzed with help of the Monte-Carlo program TRim for BackScattering (TRBS) [3]. A typical spectrum as obtained in the present study is shown in Fig. 1. Both the high-energy onset and a pronounced tail towards lower energies show the prominent contribution due to multiple scattering to the experimental yield, also at the highest particle energies. The figure illustrates the accuracy of the simulations

in reproducing the experimental results for correct choice of the scattering potential (red line for the Thomas-Fermi-Molière potential - TFM). Note, however, that in the simulations multiple scattering is strongly influenced by the exact choice of the potential (see dashed lines in the figure for the use of different screening length corrections c_a). Changes in the electronic stopping employed in the simulations are found to influence mainly the width of the spectrum (not shown) and permit thus to extract electronic stopping cross sections ϵ from a best fit to the experiment with accurate settings for the scattering potential.

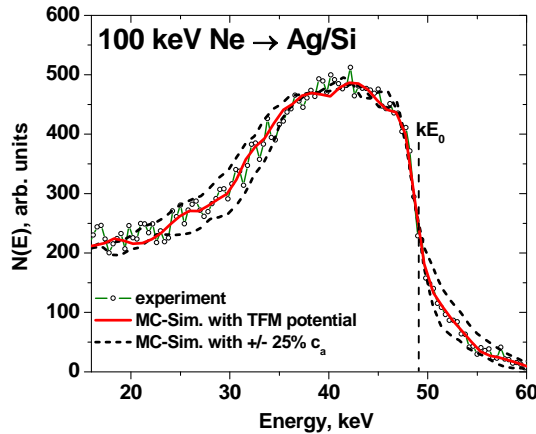


Figure 1: Energy-converted spectrum recorded for 100 keV Ne primary particles scattered from a thin film of Ag/Si (open symbols). The spectrum shape is strongly influenced by multiple scattering. The red continuous and the black dashed lines show simulated yields obtained by TRBS for the Thomas-Fermi-Molière potential without and with $\pm 25\%$ screening length correction.

In parallel to the quantification of ϵ with help of the MC-simulations, a simple evaluation within the single scattering model was performed to compare for the influence of multiple scattering and the nuclear energy loss. This comparison of different evaluation strategies was motivated by observed discrepancies between the recent results obtained in backscattering and earlier results by different groups in transmission experiments. The results of the present study show that for both, transmission and backscattering spectra, the ratio of electronic to nuclear stopping observed along the relevant trajectories significantly deviates from the tabulated values. The performed evaluation together with TRIM calculations shows that also in backscattering experiments the nuclear stopping contributions are found significantly smaller than expected along random trajectories. These results indicate an efficient trajectory selection process when evaluating spectra in typical experiments performed to extract information on the inelastic energy loss. Note, that the present results for Ne and about 100 keV primary energy also indicate that equivalent considerations can be expected to be valid for scattering of Helium at energies around or below 10 keV as well as for protons for energies around and below a few keV where similar strong multiple scattering contributions to the spectrum can be expected.

A second set of experiments was designed to study the energy loss of hydrogen and helium in a material of high relevance for the MEIS community, i.e. HfO_2 . HfO_2 was grown by atomic layer deposition on Si with a 0.5 nm thick SiO_2 buffer layer. A thorough characterization of the material system was performed in order to avoid artefacts from e.g. inaccurate stoichiometry of the oxide [4]. A combined approach via elastic resonance scattering around the resonance at 3.0375 MeV for oxygen together with channeling experiments at MeV energies was used to confirm the correct stoichiometry.

The experiments performed to measure the electronic stopping cross section ϵ [5] revealed a difference in the magnitude and the velocity dependence of the energy loss observed for H and He ions that goes beyond explanations based on the electron density of valence electrons in the system.

Figure 2 shows the observed electronic stopping cross sections normalized with the projectile velocity, yielding a friction coefficient Q according to $\epsilon = Q \cdot v$. Experimental data for protons

and helium is shown as full squares and asterisks respectively. Also shown are predictions for Q in a free electron gas (FEG) as obtained from DFT for equivalent electron density for H and He (dashed and dash-dotted lines respectively), expressed by the density parameter r_s [6]. The electron density for the shown curves is equivalent to 3.7 electrons per molecule (obtained from fitting to the data for H). Obviously, DFT-predictions based on the electron density obtained for H is not matching data for He. Instead, for helium, a density of the FEG equivalent to 11 e^- per molecule is necessary to explain the observed difference in magnitude of the electronic stopping cross section at a given ion velocity. Additionally, data for He shows less clear velocity proportionality at velocities below 1 a.u., for which DFT calculations are expected to be most applicable. Thus, even if the employed DFT-predictions for a FEG are definitely not fully applicable for a large-band insulator such as HfO_2 , the present results are a strong indication for processes different from electron-hole pair excitation contributing to the energy loss of He ions in HfO_2 .

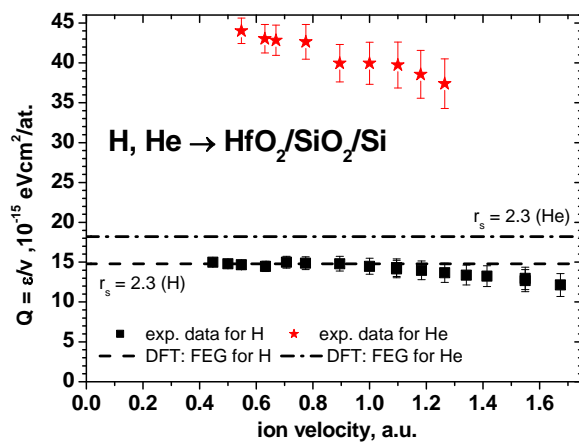


Figure 2: Electronic stopping cross sections for H and He ions in HfO_2 normalized with the projectile velocity (full squares and asterisks, respectively). Also shown are DFT predictions (dashed and dash-dotted lines).

A detailed comparison of the results with those for other materials such as SiO_2 yields further evidence for a contribution to the inelastic energy loss which is influenced by both the presence of the oxygen 2s-bands as well as the Hf 4f-states: for SiO_2 and HfO_2 , the specific energy loss of protons is found almost identical for energies where f-electrons likely do not participate in direct excitations, which is a strong indication, that energy dissipation is mainly due to excitation of O-2p-like states, which are the dominant contribution to the density of states at low binding energies in both system. Also for SiO_2 , however, He shows increased specific energy loss with respect to DFT-predictions. The effect, however, is found less pronounced which may be based in the fact, that also f-states affect the energy loss for He in HfO_2 .

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