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The role of basic energy-loss processes in layer-resolved surface investigations with ions ☆

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Abstract

Ab initio quantum mechanical calculations have been performed for the energy loss of protons backscattered from an Al surface. Results from first-order perturbation theory are compared to full numerical atomic-orbital coupled-channel calculations. It is shown that both inner shells and non-perturbative effects are important for the understanding of ion energy-loss spectra.

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1. Introduction and theoretical method

High-resolution ion energy-loss spectra may show asymmetric structures in the surface-backscattering peak. Such energy-loss distributions are influenced by the depth distribution of scattering centers, by the details of a crystalline surface structure and also by individual electronic excitation and ionization events. The details of these electronic energy-transfer processes become very important if backscattering is restricted to a few mono layers only, such as under blocking conditions.

Established atomic physics models, such as the atomic-orbital (AO) coupled-channel method have been applied to energy-transfer processes involving the inner shells of the target for different impact parameters [1]. Such large-scale quantum mechanical ab-initio calculations yield very accurate results for those cases where electrons may be described in the independent-particle picture. For atomic hydrogen these calculations are virtually exact, and thus they serve as benchmark data for simplified models. For an atomic He target [2], these calculations yield energy-loss spectra in good agreement with experimental single-collision data for different proton scattering angles. For crystalline Si targets [3] they allow to predict the channeling energy loss as function of the incident energy and of the tilt angle with respect to a main crystal axis. Here we concentrate on the proton backscattering from Al for a shadowing/blocking geometry, where very small impact parameters come into play. We go beyond a similar previous work for C and S targets [4] by application of the AO coupled-channel method instead of using the impact-parameter Born approximation (B1) that relies on first-order perturbation theory for the electronic motion.

The influence of the projectile path and of the target shell-structure on the corresponding energy-loss distributions of fast protons is considered in our calculations as will briefly be explained in the following. For each impact parameter and each electronic shell the electronic energy loss is determined either by solving the time dependent Schrödinger equation numerically (AO) or by using the computationally much less involved impact-parameter Born approximation (B1). The Al K-shell is not important for the present velocity of 100 keV, as was checked with AO calculations. For Al L-shell excitation/

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ionization, large-scale AO calculations have been performed for 400 bound and continuum eigenstates of the target with partial waves up to $l = 8$. From the final-state amplitudes resulting from these calculation the energy-loss spectra are extracted for each impact parameter and for the initial $2s_0$, $2p_0$, as well as $2p_{\pm1}$ orbitals.

Conduction/valence band excitations have been treated within B1 only, since non-perturbative effects and the corresponding energy losses are small and will not show up in current medium energy ion-scattering (MEIS) experiments. An atomic treatment of these electrons is also not problematic as long as energy loss spectra are not measured with energy resolutions below about 30 eV, where intraband transitions and plasmon excitations yield spectral structures that differ from the sum of discrete atomic transitions and the smooth atomic continuum at higher energy transfers. In fact, electronic solid-state screening and plasmon excitations cancel each other to a large extend so that the mean energy loss due to valence electrons will not critically depend on the phase of the target.

The energy transfer to a specific target atom is just the convolution of the energy-loss distributions for all electrons of this atom. The final energy loss spectrum of the protons is then given by the convolution of all atomic energy losses along the trajectory, including nuclear energy losses (specifically important for the backscattering collision). The sequence of collisions with a distribution of impact parameters is obtained from the VEGAS code for a specific scattering geometry [5].

Thus, in this work we fix the scattering geometry and derive the distribution of ion trajectories and atomic impact parameters from a Monte Carlo solution of simple two-body scattering [5]. The electronic energy losses are computed with the AO coupled-channel code for each electron that is found along the ion trajectory. Thus, we present the first ab initio results for ion-energy distributions that go beyond first-order perturbation theory.

2. Results and discussion

Actual calculations have been performed as described above for 100 keV protons backscattered from the Al(1 1 0) surface. The proton scattering-angle is 60°, the ion-entrance and ion-exit angles are also 60° with respect to the surface normal. The entrance direction [−1 0 1] corresponds to a shadow cone around all perfectly aligned bulk atoms and the exit direction [0 1 1] corresponds to a blocking direction, where backscattering from inside the bulk is strongly suppressed. Thus, first layer scattering is by far dominant for this geometry assuming an ideal crystal at low temperatures.

Fig. 1 displays proton energy-loss spectra with realistic hitting and detecting probabilities as obtained from

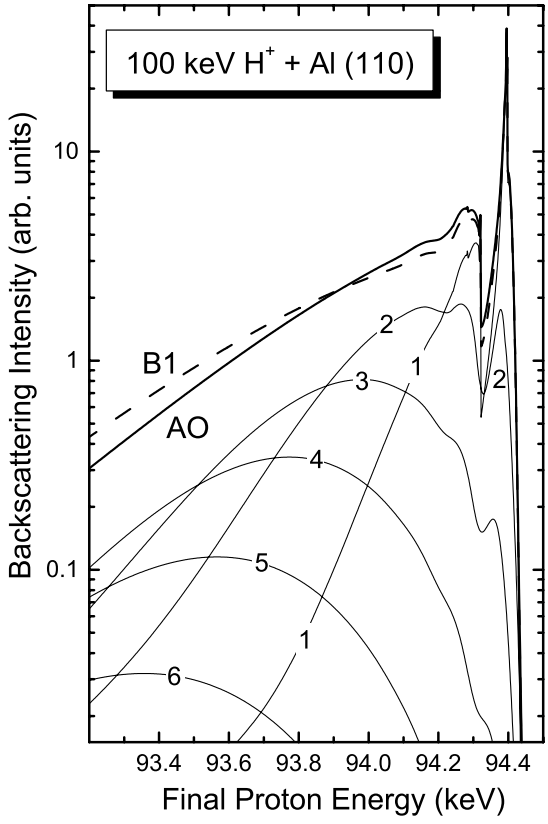

Fig. 1. Final energy distributions for protons backscattered from an Al(1 1 0) surface for a combination of shadowing and blocking directions as described in the text. Full numerical solutions of the time-dependent Schrödinger equation (AO, solid curves) are compared to results from the first-order impact-parameter Born approximation (B1, thick dashed curve). The thin solid curves labeled 1–6 show the AO contributions corresponding to backscattering from the first to the sixth layer.

the VEGAS code [5]. Fractional backscattering from up to the seventh layer is considered in the computation of the sum spectra (thick solid and dashed curves). For the electronic energy loss shown in this figure, we have simply assumed an ideal crystal structure. Thus, the impact parameters b with respect to the atomic excitation and ionization processes are replaced by $b = 0$.

At energies around 94.4 keV both calculated energy distributions (B1 and AO) show a pronounced sharp peak structure due to elastic collisions and multiple valence-band excitations. At lower energies (about 94.28 keV) a secondary asymmetric peak is clearly visible in the spectrum. This peak is due to L_1- and $L_{2,3}$-shell ionization into high-energy continuum states.

As can be seen from the fractional backscattering contributions (solid curves labeled 1–6), the asymmetry of the energy-loss spectrum close to the peaks is influenced by backscattering from deeper layers as well as by the inherent asymmetry of the energy-transfer spectrum for individual inner-shell electrons. The two dominant peak structures in the AO and B1 sum spectra are due to

backscattering from the first layer (see solid curve with label 1). At about 94.16 keV a small bump is superimposed on the low-energy wing of the AO sum curve. This bump is the only clear signature of second-layer scattering (solid curve with label 2) in the whole energy spectrum.

Comparison of the exact independent-electron results (AO) with first-order perturbation theory (B1) in Fig. 1, shows differences exceeding 40% at large energy losses. This is a clear indication for the breakdown of the B1 model, because target and projectile potentials as well as initial-state and final-state wave functions are identical for both theoretical results. The differences are related to higher-order effects, which in the present case reduce the mean energy loss. Thus, full non-perturbative calculations appear to be necessary for a quantitative comparison with experimental data, the final goal of the present work.

If we consider also thermal vibrations and surface relaxations, larger impact parameters ($b \approx 0.1$ Å) come into play for the electronic energy loss and the low-energy wing of the AO energy distribution is suppressed further in comparison with the solid curve in Fig. 1 [6]. After consideration of the experimental resolution, this brings our ab initio results in reasonable agreement with experimental data as will be shown in a subsequent publication [6].

3. Conclusions

In this work, we present and discuss the first accurate solutions of the time-dependent Schrödinger equation for MEIS. We apply large-scale AO coupled-channel calculations for 100 keV protons backscattered from Al(1 1 0) for a shadowing/blocking geometry. It is shown that the target L-shell is mainly responsible for asymmetric energy-loss distributions and a significant asymmetry is related even to backscattering from the topmost layer only. Thus, an interpretation of asymmetric peak shapes cannot directly be related to the depth distribution of backscattering events without a profound knowledge on the individual electronic energy losses. Dependent on the target elemental composition and the corresponding shell structure, even pronounced secondary peaks from the first layer are conceivable that may be misinterpreted as backscattering from deeper layers. As is also shown in this work, quantitative results call for advanced collision models and simple scaling rules or even first-order perturbation theory yield just rough estimates.

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