Energy loss of 100-keV hydrogen atoms during grazing scattering from Cu(111)

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The energy loss of 100-keV hydrogen atoms scattered under a grazing angle of incidence from a clean and flat Cu(111) surface is studied in coincidence with the number of emitted electrons. From the dependence of the energy loss on the angle of incidence we deduce the position dependent stopping power which shows the same functional dependence but a factor of about 2 higher values than recent calculations. From the number of emitted electrons related to a specific energy loss we reveal processes related to electron emission as dominant channel for energy dissipation of scattered projectiles.

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When energetic atomic particles interact with matter, part of their kinetic energy is dissipated into excitations of target and projectile. For metals these interactions comprise excitations of single electrons as well as collective excitations of the solid [1]. Local excitations of electrons lead to the formation of electron hole pairs and emission of electrons, whereas collective excitations result in the production of plasmons. Representative for numerous experimental and theoretical studies on the energy loss of fast ions in metals, we refer to a detailed discussion on this topic by Ritchie *et al.* [2].

An important experimental development concerning studies on electronic stopping is the scattering of fast atoms or ions from a surface under a grazing angle of incidence [3] where the projectiles are reflected in front of the surface plane in terms of defined trajectories. In this regime of surface channeling [4], contributions of energy transfer to lattice atoms ("nuclear stopping") is negligible [5] so that for metal targets only electronic stopping is present. Kimura *et al.* [6] proposed to make use of this feature for studies on the interactions of the projectiles with the electron gas in the selvedge of the solid-vacuum interface. From the controlled variation of projectile trajectories in this collision regime valuable information on the energy dissipation processes can be derived. The analysis of experiments in terms of position dependent stopping powers provide data to test theoretical model calculations [7].

Recently Alducin *et al.* [8] presented a detailed theoretical study on electronic stopping of protons in front of a Cu(111) surface. These authors have investigated the effect of the electronic band structure of a metal target on the energy loss of projectiles. This can be considered as a substantial step towards a realistic description of electronic excitations induced by fast atomic projectiles, since former studies used the jellium approximation for calculations of the properties of conduction band electrons. In this respect, the Cu(111) surface is an attractive system, since this face of a Cu crystal shows a pronounced projected band gap normal to the surface and has an occupied surface state with a binding energy of −5.3 eV [9]. The electronic structure of this surface can be described theoretically on the basis of a model potential that shows a modulation inside the metal and merges asymptotically to a classical image potential in front of the metal [10]. The main motivation of our work is a quantitative experimental test on the theoretical predictions given in Ref. [8]. We have furthermore recorded energy loss spectra in coincidence with the emission of a defined number of electrons and can relate in detail the overall dissipation of projectile energy to the ejection of electrons into vacuum.

In our experiments we have scattered 100 keV neutral H atoms from a Cu(111) surface under grazing angles of incidence of typically one degree. Neutral H atoms were used instead of protons in order to avoid effects of electric fields from the electron detector on the trajectories of incident projectiles. Previous studies have shown that stopping of light ions in this velocity regime is independent of projectile charge [11]. The target was prepared via cycles of grazing sputtering with 25-keV Ar^+ ions and subsequent annealing. This procedure results in atomically clean and flat monocrystalline surfaces [3]. The base pressure in our uhv setup with three differential pumping stages was some 10^{-11} mbar. The energy of incoming and scattered beams was measured with a time-of-flight (TOF) setup where the chopping of a proton beam from a small ion accelerator was achieved with sets of diaphragms and electric field plates biased with voltage pulses of about 500 V with a rise time of some ns. The chopped ion beam was then neutralized via near resonant electron transfer in a Kr gas cell. The start signal of our TOF setup was obtained from a multichannel plate (MCP) detector that was positioned 1.16 m behind the target. Above the target surface a further detector was positioned for recording the number of electrons emitted during impact of single projectiles. This detector consists of a surface barrier detector (SBD) biased to a voltage of 25 kV, and the resulting pulse height for electron bombardment is proportional to the number of emitted electrons [12]. With a bias voltage of some 100 V applied to a highly transparent grid more than 90% of emitted electrons are recorded with our setup. Relating the signal of a time-to-amplitude converter (TAC) of our TOF setup to the SBD pulse height allows us to obtain energy loss spectra for a specific number of emitted electrons.

In Fig. 1 we show an energy loss spectrum for 100-keV H atoms scattered from a Cu(111) surface under a grazing

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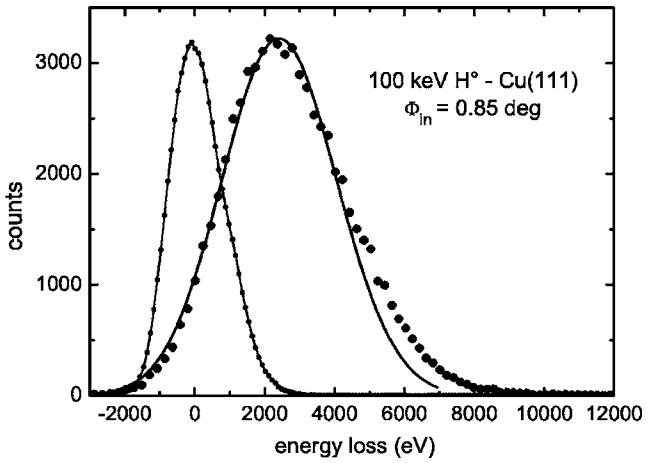

FIG. 1. Energy loss spectra for 100-keV H atoms. Dots and thin solid curve, spectrum for incident beam; full circles, spectrum for beam scattered from Cu(111) under $\Phi_{in}=0.85°$; solid curve, best fit to Gaussian line shape for convolution with energy distribution of incident beam.

angle of incidence $\Phi_{in}=0.85°$. This spectrum is derived via conversion of a TOF spectrum to an energy scale. The dots connected by a thin solid curve show the spectrum for the incident beam. Owing to the fast motion of projectiles (velocity corresponds to 2 a.u.=2 atomic units) with beam chopping in the ns domain, an overall energy resolution of about 2% (about 2 keV) is achieved with our setup. This is of the same size as the observed energy loss. However, since energy shifts can still reliably be derived from such broadened spectra, the accuracy in the experiments is sufficient with respect to the current status in the analysis of data. The full circles in Fig. 1 represent the energy spectrum of the scattered beam, which reveals a well resolved energy shift and a broadening with respect to the incident beam. The thick solid curve represents a best Gaussian fit to the data for convolution with the energy profile of the projectile beam. Aside from slight deviations between fit and data in the high energy loss tail, the data are fairly well described by a symmetric line shape, i.e., within our experimental uncertainty most probable and mean energy loss can be considered to be the same. We note that the intrinsic width of the incident beam from our small ion accelerator amounts to less than 1 keV.

The position dependent stopping power is derived from measurements of energy loss as function of angle of incidence using the concept proposed by Kimura *et al.* [6]. Full circles in Fig. 2 represent the energy loss as function of incidence angle for 100-keV H atoms scattered from Cu(111) which reveals a monotonic decrease with increasing angle. The curves shown in the figure are calculations by Alducin *et al.* [8]. These theoretical data are obtained from calculated position dependent stopping powers as plotted in Fig. 3 by integration over complete trajectories using screened interaction potentials [1]. The different types of curves in Figs. 2 and 3 correspond to the different approximations used in the calculations.

The dotted curves in Figs. 2 and 3 represent calculations within linear response theory where the band structure of

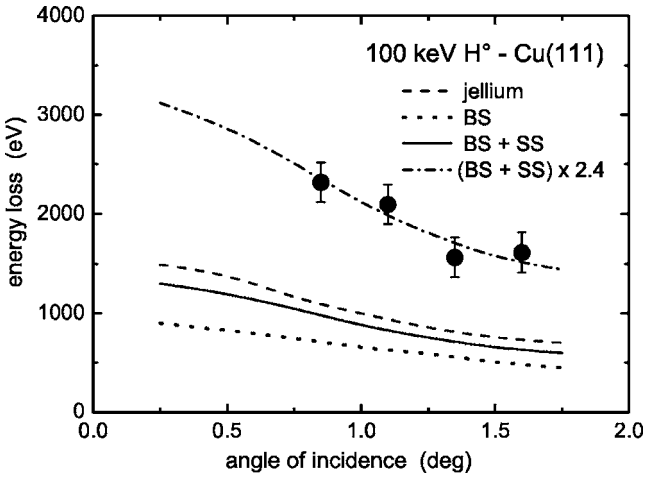

FIG. 2. Mean energy loss as function of angle of incidence for scattering of 100-keV H atoms from Cu(111). Full circles, experiment (this work); dotted curve, calculations from Ref. [8] based on band structure (BS); solid curve, calculations based on band structure and surface state (BS+SS); dashed curve, calculations based on jellium model; dashed-dotted curve, (BB+SS)×2.4. Corresponding position dependent stopping powers are given in Fig. 3.

Cu(111) is taken into account by using electronic properties derived from a realistic one-dimensional model potential normal to the surface [10]. The solid curve considers in addition contributions from the (occupied) surface state of Cu(111), and the dashed curve is obtained using the free electron model ("jellium" model) for the electronic structure of the surface. As pointed out in the original paper [8], the difference in electronic stopping power and resulting energy loss between the calculations using the realistic band structure (including surface state) and the jellium model is surprisingly small. The origin for this close coincidence is not clear at present; the theoretical results, however, indicate,

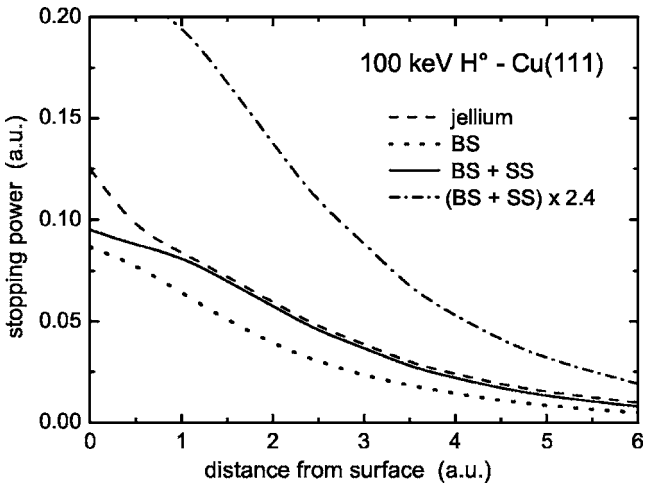

FIG. 3. Stopping power as function of distance from topmost surface layer for 100-keV H atoms in front of Cu(111) surface. Dotted curve, calculations from Ref. [8] based on band structure (BS); solid curve, calculations based on band structure and surface state (BB+SS); dashed curve, calculations based on jellium model; dashed-dotted curve, (BS+SS)×2.4.

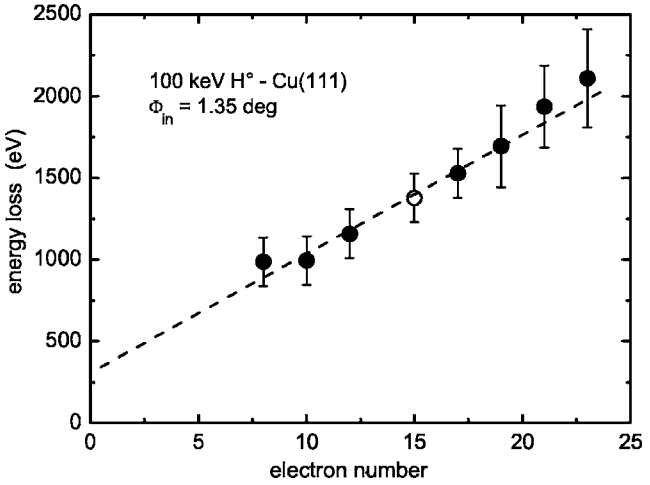

FIG. 4. Energy loss as function of number of emitted electrons for scattering of 100-keV H atoms from Cu(111) under $\Phi_{in}=1.35°$. Full and open circles represent experimental data. Open circle, energy loss for emission of $n=15$ electrons. Dashed line, linear extrapolation to energy related to emission of no electron.

that it is hardly feasible to find evidence for differences between the jellium approximation and the realistic band structure for electronic stopping in experiments. For a more detailed discussion on this topic we refer to the paper by Alducin *et al.* [8].

The experimental data for energy loss and the resulting stopping power differ clearly from the calculations. However, the angular dependence observed in the experiments agrees well with theory. In fact, multiplication of the theoretical results (band structure plus surface state) by a factor of 2.4 results in good agreement with the data. As pointed out already by the authors of the calculations, this discrepancy can be attributed, at least to a major extent, to contributions of 3*d* electrons which were neglected in the present calculations. Our measurements support such an interpretation, but a more elaborate approach beyond the simple two band model of Ref. [8] including *s*, *p*, and *d* bands has to be considered in future theoretical work on this topic.

A second part of our work concerns the coincident measurement of projectile energy loss spectra for a specific number of emitted electrons. These measurements allow us to provide an estimate on the amount of projectile energy dissipated in terms of electron emission. Details on our setup for performing TOF studies in coincidence with the number of emitted electrons can be found elsewhere [13]. Via cuts in coincident TOF spectra for specific intervals of SBD pulse heights (equivalent to emission of a defined number of electrons), we derive the projectile energy loss for a given number of emitted electrons. This energy loss for 100-keV H atoms scattered from Cu(111) under $\Phi_{in}=1.35°$ is plotted as function of electron number in Fig. 4. Irrespective of the large uncertainties in our present data caused by the limited time (energy) resolution of our setup, we reveal a substantial increase of the energy loss with increasing electron number. In passing we note such a type of analysis has been performed recently by Stöckl *et al.* [14] for scattering of multiply charged Ar ions from LiF(001) at keV energies.

The increase of energy loss with electron number as displayed in Fig. 4 results in an energy of about 60 eV per emitted electron. We measured a total electron yield of about 15 electrons (data point for $n=15$ is marked by open symbol) so that in average a total projectile energy of about 900 eV is dissipated in a process related to electron emission. This is more than 2/3 of the overall energy loss. From a linear extrapolation of the energy loss shown in the plot in Fig. 4 we reveal that an energy loss of less than 500 eV is related to the emission of no electron. This amount of energy is attributed to excitation processes in the target without emission of electrons into vacuum [collective excitations of surface and bulk plasmons with low probabilities for emission of electrons, local excitations with energy transfer to electrons less than the target work function of about 5 eV for Cu(111)]. From our measurements we conclude that those collective internal excitations of the target play only a minor role here compared to local electron—hole pair excitations induced by the fast projectile.

These local excitations can be considered as binary collisions of atomic projectiles with conduction electrons. The energy transfer in head-on collisions to an electron initially at rest amounts to about 220 eV for the projectile velocity of our experiments $v_{proj}=2$ a.u. For a Fermi distribution of conduction electrons of Cu with Fermi momentum $k_F \approx 0.7$ a.u., the maximum energy transfer to electrons (in atomic units) is $(2v_{proj}+k_F)^2/2 \approx 300$ eV [15]. A detailed analysis of the electron emission process is beyond the scope of the present paper, since electron transport in the target and crossing of the solid-vacuum interface will affect electron emission in a decisive manner [16]. However, a crude estimate on a mean energy loss per local excitation event under the assumption that about one half of the excited electrons will reach vacuum, is consistent with the observed mean energy loss of about 60 eV per ejected electron.

In conclusion, motivated by recent theoretical work [8] we have performed an experimental study on the energy loss of fast hydrogen atoms scattered from a Cu(111) surface under grazing angles of incidence, in order to test the effect of a realistic electronic structure of a metal on stopping. We find stopping powers that are in fair agreement with the calculated dependence for the distance from the surface. However, the experimental values are generally a factor of about 2.4 higher than theory. Following the argument already given by the authors of Ref. [8], we suggest the inclusion of 3*d* electrons in the calculations. This is expected to improve the agreement with the data considerably. We hope that our work will stimulate further work on this problem, also in view of data obtained by the additional detection of emitted electrons. Since a substantial part of the projectile energy loss is converted to emitted electrons, it would be of basic interest to explore this feature in the stopping process in more detail.

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