

Inelastic energy loss of medium energy H and He ions in Au and Pt: Deviations from velocity proportionality

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(Received 27 April 2012; revised manuscript received 23 July 2012; published 4 September 2012)

Electronic energy loss of light ions in nm films of Au and Pt has been studied at keV ion energies. For H the electronic stopping power S is found to exhibit the expected velocity proportionality at low ion velocities, which confirms the anticipated excitation mechanisms responsible for the energy transfer between ions and the electrons in the solid. In contrast, for He, S shows a clear deviation from velocity proportionality for both materials at ion velocities below 0.8 atomic units, i.e., 64 keV. This result indicates a change in the interaction mechanisms active at the investigated ion velocities, which cannot exclusively be interpreted from the density of states of the target. Instead, the more complex electronic structure of the He ion enables an additional energy loss channel due to charge exchange by atomic level promotion. Associated energy losses together with a changed equilibrium charge state permit an explanation of the observed phenomenon.

DOI: [10.1103/PhysRevB.86.094102](https://doi.org/10.1103/PhysRevB.86.094102)

PACS number(s): 61.82.Bg, 68.49.Sf, 34.50.Bw, 34.70.+e

I. INTRODUCTION

When an ion moves in matter it inevitably interacts with the electrons in the target material. This interaction typically leads to excitation of the electronic system due to an energy transfer between ion and target electrons and hence a deceleration of the ion. The underlying mechanisms are of fundamental interest, and furthermore the specific energy loss of keV ions in solids is of importance for many fields of research such as astrophysics, plasma physics, and material science.¹⁻³

An ion that moves with velocity v in a medium is subject to a decelerating force, which is commonly denoted as stopping power S . To avoid a density dependence of S , the electronic energy loss is frequently quantified by the stopping cross section (SCS) ε , which is closely related to S via $\varepsilon = (1/n) \cdot S$, with the atomic number density n .

Electronic stopping of fast ions, i.e., with energies of several mega-electron-volts per nucleon, has been of interest for theoretical studies since the early days of ion physics.⁴ At sufficiently high energies the energy transfer between the projectile and the electronic system of the target is mainly due to excitation of core shell electrons; thus, details in the density of states of the irradiated substance have negligible influence. In this regime the ions are typically fully stripped, which facilitates the theoretical treatment. With decreasing ion energies the projectile may bind electrons, and charge state effects in electronic stopping have to be considered in order to facilitate a reasonable prediction of ion deceleration.⁵⁻⁷

The electronic energy loss is known to exhibit a noticeable dependence on the impact parameter.⁸ However, the impact parameters relevant for these electronic processes are too large to lead to a significant elastic energy loss in the atomic scattering at high energies. Therefore, no interrelation between electronic and nuclear losses has to be expected. In the regime of medium-energy ion scattering (~ 100 keV), however, backscattering collisions are correlated with an increased probability of inner shell ionization.^{9,10}

The maximum possible energy transfer decreases with decreasing ion energy. Consequently, the structure of the valence and conduction band of the solid becomes more and

more relevant and eventually dominates the physics behind the electronic energy loss of ions with keV kinetic energy. Light ions are of special interest in order to obtain an understanding of the excitation of the solid and the associated energy loss processes of the ions for two reasons. First, their own electronic structure is simple and permits the focus to be on the excitations in the target material. Second, elastic energy losses are still comparatively small when dealing with energies in the range of ~ 10 – 200 keV.^{11,12} In this regime the projectiles are considered to interact almost exclusively with the delocalized electrons of the target material.

Consequently, a commonly employed model system with respect to the electronic energy loss of slow ions, particularly with regard to metals, is that of free electron gas (FEG). Fermi and Teller¹³ and consecutive studies¹⁴⁻¹⁶ predicted for this case that ε is proportional to the ion velocity, v . This has indeed been experimentally observed for protons at $v < v_0 \equiv c/137$ in many materials.¹⁷ For noble metals, however, a different velocity scaling of the electronic energy loss of slow protons has been found. In Au a significant deviation from velocity proportionality towards lower values was observed at ion velocities below 0.6 a.u.¹⁸ Subsequent studies revealed a similar effect for H in Cu and Ag and explained the observations by a contribution of “nearly free d electrons” to the energy loss at $v > 0.6$ a.u., which starts to diminish for lower ion velocities.¹⁹ Comparable phenomena are known for systems with an excitation threshold for electron-hole pair creation, e.g., noble gases or band gap materials.²⁰⁻²³ For all noble metals at proton velocities ~ 0.2 a.u., S is observed to become velocity proportional again. The velocity for which the transition between different regimes is observed depends on the binding energy of the d states,²⁴⁻²⁶ which gives confidence in the interpretation of the velocity scaling of ε for protons due to the nontrivial density of states in noble metals.

In principle, all observed phenomena are expected to be also valid for He ions. Indeed, a change in velocity scaling at very low ion energies in Cu and Au was observed.²⁵ However, the absolute values of ε for He at very low ion velocities—corresponding to a small number of electrons

that can be excited—were typically found much higher than predicted for a FEG with low density. Very recently, the theory succeeded in a quantitative prediction of the velocity scaling of electronic stopping for very low-energy H and He ions in Au, particularly addressing the long-standing problem of the stopping power ratio between H and He.²⁷ The important role of d electrons in the energy loss for very slow ions is further strengthened by this study.

However, for some materials like Mg or SiO₂ the experimentally deduced SCS for protons and He could not be explained on a common basis with the aforementioned results.^{21,28} A recent study revealed that also for Al, which is expected to closely resemble the model system of a FEG, data obtained for protons perfectly match the expectations, whereas for He a distinct deviation from velocity proportionality was observed.²⁹ The origin of the effect can be explained by the more complex structure of the He projectile and interactions that go beyond what is included in a FEG model: additional contributions to the energy loss have to be expected because of the possibility of charge exchange in close collisions between projectile and target nuclei. In the reionization event energy will be transferred to the subsequently lost electron due to atomic level promotion. The relevant quantities, i.e., the shift of the occupied He-1s level, show a strong dependence on the interaction distance.^{30,31} Consequently, the probabilities for the process to happen along a trajectory with insignificant deflection, i.e., elastic energy losses, are expected to be energy dependent. A common feature of the materials for which the velocity scaling of electronic stopping for He ions cannot be interpreted exclusively from the electronic structure of the target is a strong shift of the He-1s level at comparatively large interaction distances.³² Thus, the effect can be observed at very low ion energies and, due to decreasing ε , can significantly contribute to the energy loss.

In light of these findings it was planned to investigate the interaction of H and He ions in Au and Pt at velocities $0.5 < v < 1.5$ in a comparative way. The specific target elements were chosen because they both feature a similarly structured density of states: the d states are, however, shifted by about 2 eV^{33,34} with lower binding energy for Pt. This difference in electronic structure is expected to lead to significantly different velocity scaling of ε .

In parallel, the systems of He scattered from either Au or Pt are known to feature a rather similar energy dependence of charge exchange processes with He ions in low-energy ion scattering. However, these dependencies are very different from that known for He and Al. First, the energy thresholds for a reionization event in given scattering geometry are about five to six times higher.³² Second, neutralization processes for He⁺ in Al are by far more effective than, e.g., in Au.^{35,36}

The present study was performed to answer the following two questions. Is electronic energy loss of light ions with several keV kinetic energy sufficiently well described by the interaction of the ion charge with a modified FEG, including excitation thresholds for certain electronic states? If this is not the case, does the more complex electronic structure of He require a more detailed description including other energy loss mechanisms in order to explain the experimental observations?

II. EXPERIMENT

The experiments to deduce electronic SCSs ε were performed using the time-of-flight medium-energy ion scattering (TOF-MEIS) setup at Uppsala University.³⁷ Atomic and molecular ions with primary energies E_0 in the range of 25–170 keV were employed: Molecular ions (H₂⁺ and D₂⁺) extend the range of accessible energies to $E_0/2$, where vicinage effects³⁸ are negligible.³⁹

Sets of thin films of amorphous Au and Pt were evaporated in different thicknesses on top of cleaned Si wafers using a high-fluence evaporation system. After evaporation, a set of samples was characterized by Rutherford Backscattering Spectrometry (RBS) using two MeV He⁺ ions as primary particles. RBS spectra were recorded at different tilt angles to permit accurate thickness determination by different independent methods. Relative measurements with respect to a thick reference sample, evaluation of the spectrum width, and normalization by the Si substrate signal yield concordant results and altogether yield a final thickness precision of 2% mainly due to the experiments statistics. Nevertheless, an eventual error in employed stopping powers might introduce an error in the accuracy, which is due to the high employed energies expected to be below 2%. Evaluation of the RBS spectra was performed using SIMNRA.⁴⁰ The resulting thicknesses of the Au films employed for the energy loss experiments were 169 Å, 251 Å, and 426 Å. The thickness of the Pt film was 164 Å.

Spectra of backscattered H and He projectiles were obtained by TOF-MEIS. TOF-MEIS permits one to obtain spectra at extremely low particle doses so that virtually no alteration in sample thickness and morphology can occur during a measurement. Furthermore, particles can be detected irrespective of their final charge state. Evaluation of energy-converted experimental spectra was performed using the TRim for BackScattering (TRBS) simulation code.⁴¹ This methodology was employed to account for the increasing influence of multiple scattering at lower energies and the consecutively increasing nuclear energy losses¹¹ as well as the changing scattering potential. Figure 1 shows a spectrum obtained for

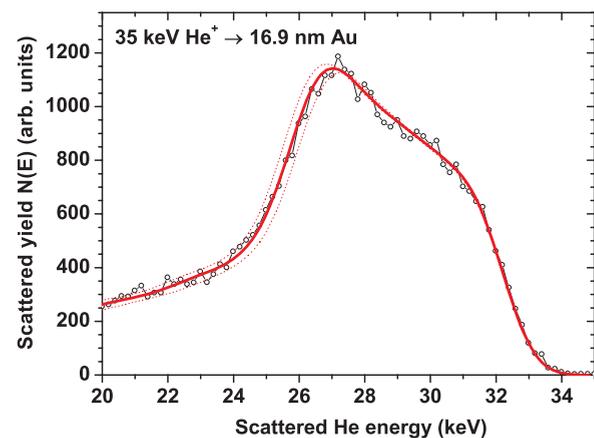


FIG. 1. (Color online) Experimental energy spectrum obtained for 35 keV He⁺ projectiles backscattered from a 169 Å Au film on Si (circles). Simulated spectra (TRBS) for different electronic stopping cross sections ε are shown as solid and dotted lines. For details see text.

primary 35 keV He ions scattered from a Au film with a thickness of 169 Å. The spectrum resembles in its main features an RBS spectrum, which indicates that the multiple large angle scattering is still of minor importance. This can also be seen from the high-energy and low-energy edges of the spectrum, which are rather similar in slope and mainly defined by the system resolution. The shape of the plateau is characterized by the energy dependence of scattering cross section and ε . The width of the spectrum is, for a given scattering geometry, determined by the energy loss in the film. Spectra simulated by TRBS with adjusted electronic energy loss to fit the spectrum width are presented in Fig. 1. For the optimal choice of ε the spectrum is perfectly reproduced (full line); a change in ε by only 3.5% gives a significantly worse fit (dashed lines), yielding a very small contribution to the error for the deduced SCS. Considering all factors, the total error is expected to be below 3%, mainly due to the RBS calibration. Note that a possible error in sample thickness calibration will only introduce a scaling factor and thus not influence the velocity scaling of deduced SCS.

III. RESULTS

Electronic SCSs were deduced for H and He in Au and Pt for ion velocities ranging from 0.6 to 2.6 a.u. for protons (~ 10 to 170 keV/amu) and 0.5 to 1.2 a.u. for He (25 to 150 keV). For Au, targets with three different thicknesses were employed and yielded equivalent results for ε . Figure 2 presents the experimentally obtained SCS for H and He in Au as a function of the projectile velocity (filled symbols). The figure also holds two other experimental datasets (open symbols) for similar energies, as investigated in the present study,^{42,43} as well as low-energy data for H and He.²⁵ Also shown are density functional theory (DFT) predictions (dashed and dotted lines for H and He, respectively) employing a FEG

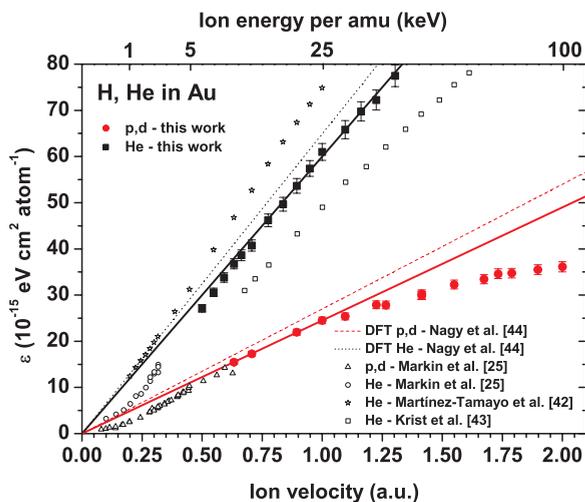


FIG. 2. (Color online) Electronic stopping cross section for H and He ions in Au as a function of projectile velocity and ion energy. Also shown are datasets for H and He for low ion energies from Ref. 25 and two datasets for He that cover a similar energy regime as the present investigation (see Refs. 42 and 43). Predictions from DFT for H and He in a FEG are shown as dashed and dotted lines, respectively (see Ref. 44). The solid lines are to guide the eye.

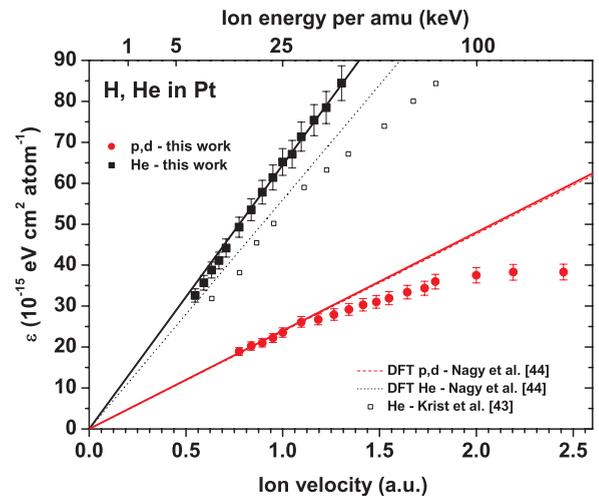


FIG. 3. (Color online) Electronic stopping cross section obtained for H and He ions in Pt as a function of projectile velocity and ion energy. Also shown is data from Krist *et al.* (Ref. 43) for He. Predictions from DFT for H and He in a FEG are shown as dashed and dotted lines, respectively (Ref. 44). The solid lines are to guide the eye.

model⁴⁴ using values for the density parameter r_s of the FEG from Ref. 45. Data obtained for protons show perfect velocity scaling at lower energies and very good quantitative agreement with the low-energy dataset. DFT only slightly overestimates the obtained SCS. At higher energies approaching the stopping maximum, data deviate from velocity proportionality towards lower values.

Data for He show velocity proportionality starting at the highest investigated ion energies but begin to significantly deviate from $\varepsilon \propto v$ towards lower values at $v \sim 0.8$ a.u. Note that in light of the present findings, a statistically significant deviation from velocity proportionality can also be claimed for the two other experimental datasets for He in Au (see Interpretation section). For He, predictions from DFT slightly overestimate the experimentally observed values of ε .

The electronic SCSs for H and He ions in Pt are presented as filled symbols in Fig. 3. The figure also shows data from Krist *et al.*⁴³ (open symbols) as well as DFT predictions⁴⁴ (dashed and dotted lines for H and He, respectively). As with Au, data for protons exhibit perfect velocity proportionality at low energies, now perfectly matching expectations from DFT. Also in the case of Pt, data for He show a deviation from velocity proportionality at low ion velocities. The deviation is, within experimental uncertainties, observed at similar velocities as for Au, i.e., around $v = 0.8$ a.u. DFT is found to slightly underestimate data for He in Pt.

IV. INTERPRETATION

Data for protons are in excellent accordance with the velocity scaling observed in previous experiments and for the energies where this is anticipated. In the velocity range of interest (i.e., around and below v_0), the magnitude of the deduced cross sections is in very good agreement with both DFT predictions and extrapolation from low-energy experiments. This proves that a reasonable description of the

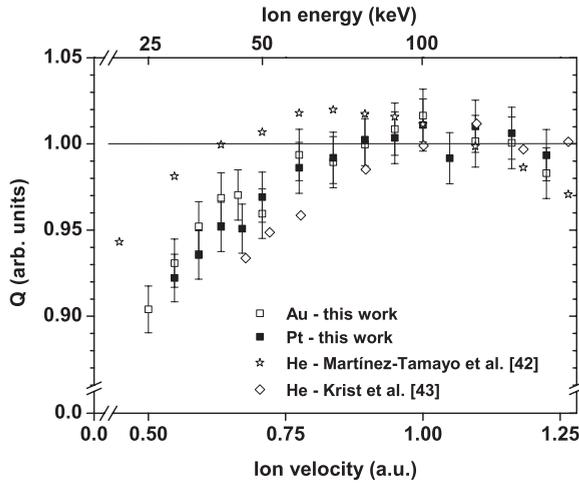


FIG. 4. Normalized friction coefficient Q assuming velocity proportionality of the electronic stopping cross sections derived from the present experiment for He in Au and Pt (open and filled squares, respectively). Also shown are the scaled data from Refs. 42 and 43 for Au (asterisks and diamonds); both also show a nonlinear velocity scaling.

energy loss of protons in the investigated metals is possible by the interaction of a point charge with the delocalized electrons of the target. In the presented velocity range no distinct excitation limits as deduced from the electronic structure are expected,^{19,42} which is compatible with the fact that $\varepsilon \propto v$ is observed. At higher energies, i.e., for $v \gg v_0$, data start to deviate from velocity proportionality. This is in accordance with the existence of a stopping power maximum at the respective energies.

In contrast with protons, deduced SCS for He exhibits a deviation from velocity proportionality at low velocities. Figure 4 presents data for the normalized friction coefficient Q , assuming $\varepsilon = Q \cdot v$ for fits to the experimental datasets for He in Au and Pt. Note that this representation is different from the common practice of plotting the stopping power ratio $R = S_{\text{He}}/S_{\text{H}}$. The employed presentation limits the observed effects exclusively to the properties of the He ion/atom and the penetrated material. In fact, a statistically significant decrease in Q , i.e., a deviation from velocity proportionality towards lower SCSs, is observed for all presented data, which start to be noticeable at velocities of about 0.8 a.u. corresponding to 64 keV He ions. In contrast, a prominent change in R was observed for Au at $v = 0.3$ a.u.⁴² Note that in this normalized representation a potential systematic error from the thickness calibration from RBS cancels out, and thus only a statistical error from the fitting procedure and the statistic of the MEIS experiments remain. This is, however, significantly smaller compared with the errors in Figs. 2 and 3, as can be estimated from the scatter of deduced data.

The observed phenomenon can be discussed in context with previous observations and the interpretation of data employed for low-energy ions. For He and Au, a deviation from velocity proportionality can be anticipated using the concepts established for protons in Ref. 19. However, in such a case, the transition should be observed at the same ion velocities, starting from $v = 0.6$ a.u., i.e., at almost half the ion energy

at which the deviation from $\varepsilon \propto v$ is observed in the present study. Note that this does not rule out that the effects observed in Ref. 19 for protons are also present for He. The observed velocity scaling is, however, an indication that another effect also plays a prominent role at these velocities. The onset of the expected deviation due to a changing number of electrons available for excitation by the projectile ions at $v < 0.6$ a.u. might in fact be partially masked by the present deviation from $\varepsilon \propto v$.

As a matter of fact the effect is also observed for Pt. For Pt, however, d bands are extending up to the Fermi level. Thus a much smaller discrepancy between number of valence electrons and effective number of electrons is found.⁴⁵ According to Valdes *et al.*,¹⁹ this is expected to lead to an almost unnoticeable deviation from velocity proportionality down to $v = 0$. Furthermore, due to the considered excitation mechanisms, a possible residual effect should be shifted to lower velocities, with the dependence on the binding energy demonstrated by Cantero *et al.*²⁶ In fact, the observed deviation for Pt has the same magnitude as observed for Au and is present for the very same ion velocity (see also Fig. 4). From the equivalent velocity scaling of ε for He in two elements with clear differences in the details of the electronic structure, it can be concluded that the observed velocity scaling cannot be attributed to the band structure of the solid.

In contrast, it can be linked to the electronic properties of the projectile. When the present results are compared to the recently discovered deviation from velocity proportionality for He ions in Al, an interpretation by charge state fluctuations under different circumstances is reasonable. For Al, a deviation from velocity proportionality for ε at ion energies below 10 keV was observed. As stated in the Introduction, for charge-changing collisions due to the promotion of the He-1s level in close encounters of He with Au and Pt nuclei, the energy threshold is much higher than for Al. Consequently, both Pt and Au are expected to show a related effect at almost equal ion energies since their threshold energies are virtually identical.

Note, however, that the induced energy dissipation process as a consequence of charge exchange will have a different character than for He in Al due to the changed energy regime and the much higher atomic number of the target atoms. A key difference is found in the changed scattering kinematics. When scattering He from Al, significant elastic energy losses, even in small angle scattering events, are found. In contrast, for Au and Pt, elastic energy losses in small angle collisions are much smaller. Furthermore, in the present study single scattering by a large angle is still dominant at all investigated energies, which is also obvious from the spectrum shape (see Fig. 1). To summarize these factors, in contrast with the system of He in Al, a less strong coupling between elastic and inelastic losses has to be expected.

Another important difference in the processes observed for He in Al is that at the energies in the present investigation the projectiles are much faster and, if they have been reionized in a close encounter with a nucleus in the target, will travel a much longer distance in the solid before they are neutralized again. This effect is even enhanced by differences in the neutralization behavior for the different target materials. Accordingly, in this energy regime the charge exchange events resulting from the He-1s level shift introduce an increased probability of the

positive charge state for He along the trajectory. Consequently, the onset of charge exchange processes along rather straight trajectories not only lead to a local energy loss due to the energy transferred to the electron, which is subsequently lost, but also to much higher ionization along the path of the projectile in the solid. This argument also explains why the discussed effect can induce a larger absolute change in the SCSs than observed in the experiments for He and Al, even if the relative contribution due to very effective electron-hole pair excitation at the present energies becomes smaller.

V. CONCLUSIONS AND OUTLOOK

From the presented experimental data and their interpretation it can be concluded that the electronic energy loss of slow protons in metallic targets is very well understood. Both the absolute values and the velocity scaling of the SCS ε are predicted with high accuracy by DFT, particularly when the density of states of the target material is reasonably considered in the calculations. This is facilitated by the fact that protons can be treated as point charges.

Data for He show a velocity dependence, which cannot be understood on a mutual basis with the concepts applied to

protons. These results can be explained by charge exchange as a consequence of atomic level promotion and associated energy losses as well as changed average charge states of the projectile. The described processes are expected to be a relevant contribution to the electronic energy loss of low-energy and medium-energy ions in many systems with characteristically different magnitudes and energy dependencies. Consequently, it seems promising to include these charge-exchange mechanisms induced by close encounters between He and the nuclei in the solid into the existing models of effective charge in a FEG. To obtain a deeper understanding of the influence of the electronic structure of the projectile on the inelastic energy loss, a thorough theoretical analysis of charge state fluctuations of different origin in combination with suitable experimental studies with very low ion energies is highly desirable.

ACKNOWLEDGMENTS

The author would like to thank Pia Lånsaker and Anders Hallén for preparation of the Au and Pt samples. Helpful discussions with P. Bauer and G. Possnert are gratefully acknowledged.

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