Survival of He^+ Ions during Grazing Scattering from a Ag(111) Surface

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He^+ ions as well as neutral He atoms with keV energies are scattered under a grazing angle of incidence from a clean and atomically flat Ag(111) surface. From a comparison of ion fractions observed after scattering of He^+ ions and He atoms we find for energies below some keV small but defined fractions of ions that have survived the complete scattering event with the surface. This feature allows us to clear up the microscopic interaction scenario for Auger neutralization of He^+ ions at a Ag(111) surface. The Auger neutralization rates are 2 to 3 orders of magnitude smaller than conventional rates derived from experiments for He^+-metal systems and agree with recent calculations.

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Charge exchange phenomena of atomic particles during their interactions with a solid are important for different fields in physics and technological applications, as, e.g., particle detection, surface chemistry, plasma wall interactions, or various surface analytical tools. Dominant electron transfer processes for atoms or ions with thermal and hyperthermal energies in front of Ag(111) are (1) *resonant electron tunneling* where the energy of the active electron is conserved and (2) *Auger neutralization* (AN), a nonresonant tunneling process with the excess energy transferred to a second electron of the solid (see, e.g., [1]).

Over the last few years, considerable progress has been achieved in the microscopic description of these fundamental mechanisms of charge transfer. Specific problems in the theoretical treatment are related to the broken symmetry at the solid-vacuum interface which comprise, e.g., a "realistic" modeling of the atom-surface interaction potentials, affected by dielectric response and perturbation of the metal by the presence of the atom or ion. For resonant one electron tunneling, nonperturbative methods were developed as "complex scaling" [2–4], "coupled angular mode" [5], or "wave-packet propagation" [6]. These methods provide electron transition rates that allow one to describe experiments on a quantitative level, if resonant transfer dominates charge exchange, as for resonant neutralization/ionization of alkali atoms in front of the surface of simple metals [7,8].

The Auger process is important, if no electronic levels of the metal are in resonance with energies of active atomic levels as met for the neutralization of noble gas ions. For illustration, we give in the upper panel of Fig. 1 a simple energy diagram for the neutralization of a He^+ ion via the Auger process in front of a metal surface: an electron from the conduction band is captured to the atomic He $1s^2$ ground state and the energy in the process is conserved via excitation of a conduction electron which can be studied spectroscopically. This feature is made use of in *ion neutralization spectroscopy* pioneered by Hagstrum [1,9].

Already in early studies, the importance of Auger neutralization for charge transfer close to solid surfaces was revealed and a microscopic description was performed in terms of Auger transition rates, approximated by an exponential decay with distance from the surface. Calculations of Auger neutralization probabilities using a rate equation result in well defined intervals of distances from the surface, where charge transfer is effective [1,9,10]. From the shift of the binding energy of atomic levels in front of a metal surface (cf. Fig. 1)—for not too small distances approximated in terms of image charge interactions by the classical $1/4z$ dependence with respect to a reference plane ("image plane")—the effective distance for Auger neutralization can be deduced from energy shifts in the electron spectra.

Based on electron spectra for impinging hyperthermal ions (energies up to some eV), distances of typically $z_{AN} \approx 2$ to 3 a.u. (a.u. = atomic units) from the image plane (about 6 a.u. from the topmost layer of surface atoms) are derived for metal surfaces [1,9–11]. At those distances, the atomic levels and Auger electron energies are assumed to be shifted by about $1/4z_{AN} \approx 1/12$ a.u. $\approx$ 2 eV. In the microscopic scenario of the scattering process, Auger neutralization takes place on the incident part of the projectile trajectories, well beyond the distance of closest approach z_{min}, and is completed before this distance is reached (sketch in the lower panel of Fig. 1).

Calculations of Auger transition rates are an intricate problem comprising screening effects in an electron gas of nonuniform density at the solid-vacuum interface. Representative for the work performed over the last few years we mention studies by Lorente and Monreal [12] and work by Cazalilla *et al.* [13]. In summarizing the outcome of these calculations, theoretical Auger transition rates differ from those deduced from previous experiments by several orders of magnitude. Owing to comparatively small theoretical transition rates, neutralization of incident ions takes place much closer to the surface plane, i.e., for hyperthermal ions at distances

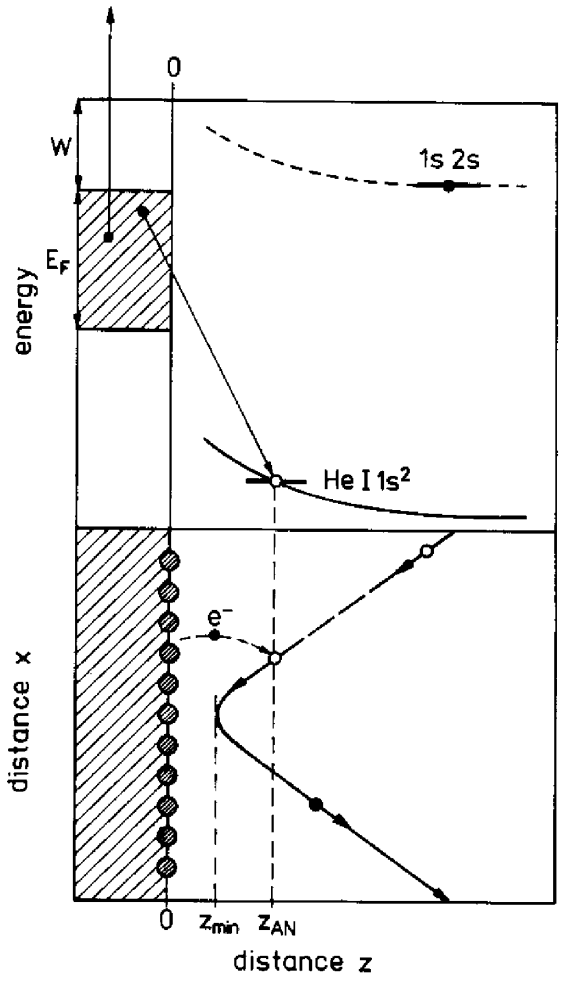

FIG. 1. Sketch for Auger neutralization of He^+ ions in front of a metal surface. Upper panel: energy diagram; lower panel: projectile trajectory.

of closest approach z_{min} or at about the jellium edge (cf. Fig. 1).

In recent papers, More *et al.* [14] and van Someren *et al.* [15] addressed this problem for grazing scattering of He^+ ions from an aluminum surface. Simulation of ion trajectories and electron spectra are consistent with the formation of atoms close to the surface, if a theoretically predicted atomic level shift reduced with respect to the classical $1/4z$ approximation is assumed [14,16]. The authors point out that also previous experiments investigating image charge effects on trajectories of ions during grazing ion scattering are consistent with those rates. In the latter method, ions are attracted on the incoming path via forces owing to their image charge, until this acceleration is ceased by their neutralization; the energy gain for the motion of ions towards the surface is deduced from the additional angular deflection of ions in comparison with neutral projectiles [17]. For the neutralization of He^+ ions in front of an Al(111) surface [18], assuming the validity of the classical $1/4z$ expression for the image potential, Auger neutralization rates comparable with former studies [1,9–11], but substantially higher than obtained by theory, and distances of neutralization well in front of the surface (about 3 a.u. from image plane) were deduced.

This brings us to the key issue of the present Letter. In the microscopic understanding for Auger neutralization, the available experimental data can be explained in two alternative ways: (1) "higher" Auger transition rates resulting in distances of neutralization at a distance from the surface, where response effects, leading to atomic level shifts and acceleration of ions, are described by the classical asymptotic limit, or (2) "lower" Auger transition rates and neutralization much closer to the surface and a reduced dielectric response at those distances compared to a $1/4z$ behavior.

From the two different scenarios, one could possibly follow the argument that alternative (2) based on theoretical input for the He^+-Al model system should be favored though in contrast to the established understanding [14,15]. On the basis of the available experimental data, however, such conclusions are not definite.

A closer inspection of the problem reveals the possibility to resolve this ambiguity by making use of a specific aspect of the neutralization process. This feature is related to fractions of ions that survive the complete scattering event with the surface. For large angle ion scattering with energies for the normal motion of several 100 eV and keV, those fractions can be substantial and affected by backscattering in close binary encounters with surface atoms leading to additional channels for charge transfer (resonant processes or reionization) [19]. For grazing incidence, scattering of keV ions proceeds in the (surface) channeling regime [20] with an energy for the normal motion in the eV domain and relatively long interaction times with the solid. Then neutralization of ions has to proceed close to the turning point of trajectories in order to detect surviving ions. The presence of ions in the scattered beam provides direct information on typical distances for neutralization and the magnitude of transition rates. For scenario (1) fractions of surviving ions are fully negligible (several tens of orders of magnitude from the incident ion fractions), whereas for (2) one might expect fractions of some 10^{-4} [21].

A crucial problem in experimental tests on this feature is the reliable detection of small ion fractions, primarily related to defined scattering conditions (very clean and flat target) so that reionization of atoms by surface imperfections is on a negligible level. In our experiments, well collimated beams of keV He^+ ions and He atoms (neutralization in gas target mounted in beam line) from a small ion accelerator are scattered at a base pressure of some 10^{-11} mbar from a Ag(111) surface under grazing angles of incidence of typically $\Phi_{in} = 1°$ to $2°$. An atomically clean and flat target surface was achieved by a major number of preparation cycles via grazing sputtering with 25 keV Ar^+ ions and subsequent annealing to about 520 °C (overall time of preparation about two

months). Angular distributions and charge fractions of scattered projectiles are recorded either (1) by means of a channeltron detector with an entrance aperture covered by an ultrathin carbon foil in order to achieve an equal detection efficiency for scattered particles of different charge (He^0 and He^+ here) or (2) by a position sensitive channel plate operated via a delay line. For our studies, both detectors have their specific benefits: the channeltron with foil allows one to measure reliable charge fractions, whereas the channel plate provides with high detection efficiency data on the complete angular distributions of scattered projectiles and thus on angular shifts owing to image charge effects on the ingoing and outgoing trajectories.

In Fig. 2 we display ion fractions as a function of projectile energy for incident He^+ ions (full symbols) and for He^0 atoms (open symbols) scattered from Ag(111) under $\Phi_{in} = 1.35°$. For neutral projectiles we observe within our detection limit (about 10^{-5}) no ion fractions for energies below 5 keV, then a monotonic increase with projectile energy takes place. A similar increase is also found with He^+ projectiles, which points towards an equilibrium of final charge states via electron capture and loss, where the electron loss may proceed via a kinematically assisted Auger ionization mechanism [22].

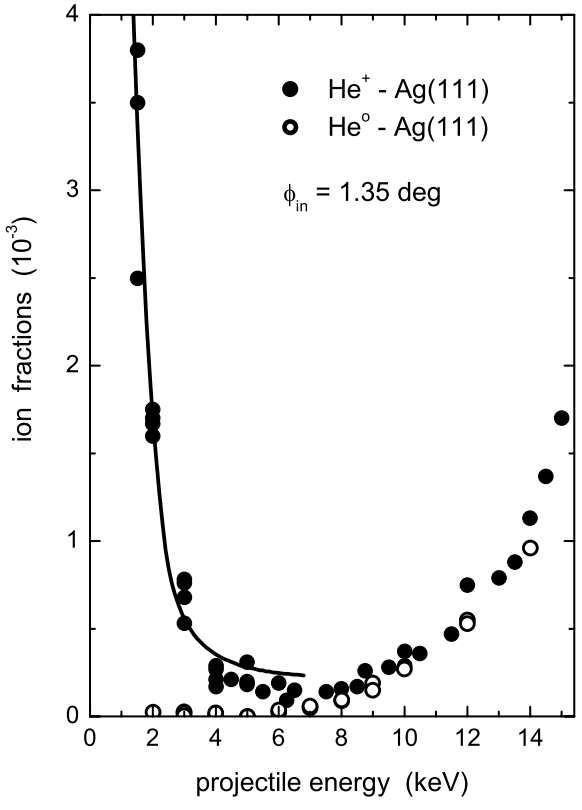

FIG. 2. He^+ ion fractions in scattered beam for He^+ (full circles) and He^0 projectiles (open circles) as a function of projectile energy. Solid curve: best fit of Auger neutralization rates to rate equation approach outlined in text.

The striking feature of our data is small ion fractions in the scattered beams for incident He^+ ions. These fractions reach a broad minimum of some 10^{-4} between about 4 and 8 keV, presumably the detection limit of previous studies. The defined difference to fractions observed for neutral projectiles shows that contributions of ionization caused by imperfections of the target surface are on a negligible level. New and unexpected came the finding that for further reduced projectile energies the ion fractions show a pronounced increase to several per mille, which is not seen with neutral projectiles.

The survival of small fractions of ions from the complete scattering event is a clear indication that Auger neutralization has to proceed not too far from the turning point of the trajectory. In a more specific analysis we performed computer simulations of classical projectile trajectories by solving the equation of motion for surface channeling conditions by making use of averaged universal interatomic potentials [23]; for the image potential we considered asymptotically a $1/4z$ behavior, at closer distances reduced values as calculated for the He-Al system by Merino *et al.* [16]. The solid curve in Fig. 2 represents results from the simulations, where the occupation of ions is obtained from a master equation approach taking into account the population of the He $1s^2$ ground state of the neutral atom only (populations of excited states via resonant neutralization are expected to play no role here, because of the work function of Ag(111), $W = 4.65$ eV). In the calculations the Auger neutralization rate is approximated by the exponential decay $W = W_o \exp(-z/z_A)$ with parameters W_o, z_A, and z_o being the distance where W saturates [$W = W(z_o)$ for $z \leq z_o$]. The final ion fractions depend sensitively on these parameters which allows us to fix those in best fits to data.

In Fig. 3 we show resulting Auger neutralization rates for the He^+-Ag system as a function of the distance from the topmost surface layer [dashed curve (1), thick solid part indicates interval of distances probed by the experiment]. Since detailed calculations for our system are not known to us, we compare with recent theory for He^+-Al (symbols) and find rates of the same order of magnitude. In view of the different electronic structures, the good agreement of the transition rates for the different systems should not be overestimated. However, we stress that rates derived here are about 3 orders of magnitude smaller than those obtained in the past from the analysis of experimental data for He^+-Al and other metal targets [dashed curve (2)]. The presence of small fractions of surviving ions can be understood only by assuming Auger neutralization rates which are comparable to those obtained by theory over the last few years [12–14].

From our simulations we reveal a typical distance of minimum approach $z_{min} \simeq 2$ to 2.5 a.u. and a mean distance for neutralization $z_{AN} \approx 3$ a.u.; i.e., Auger neutralization takes place close to the jellium edge and image

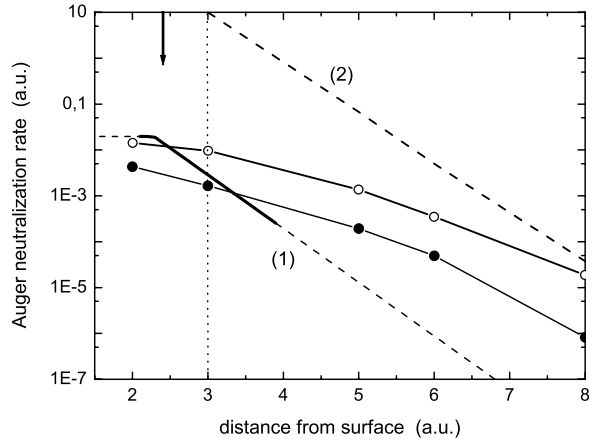

FIG. 3. Auger neutralization rates in a.u. as a function of distance from the surface. Thick and dashed curve (1): this work; dashed curve (2): analysis of experiment for He^+-Al(111) by Hecht *et al.*; open and full circles: calculations by Cazalilla *et al.* for He^+-Al with perturbed and unperturbed surface by the presence of an ion. Vertical dotted line: position of image plane; vertical arrow typical distance of closest approach.

plane. In our simulations we reveal that the accuracy of the resulting rates is primarily limited by the knowledge of the interaction potential. This potential affects the distance of closest approach $z_{\min}$ and leads to a corresponding parallel shift of the AN rates with distance from the surface; the uncertainty of this shift is estimated to about 0.5 a.u. However, the absolute values of rates are unchanged, since these directly determine the fractions of survived ions.

Image charge effects on trajectories for grazing ion surface scattering make measurements of charge fractions, in particular, at low projectiles energies, a nontrivial task. In our experiments we observe distributions for incident He^+ ions shifted towards larger angles of scattering in comparison to those for neutral projectiles. From the angular shift we derive a gain for the mean normal energy for ions of about 1.3 eV.

Applying the concept of classical image charge one would derive from this energy gain distances for AN of about 6 a.u. from the image plane (about 9 a.u. from the surface), so that transition rates would be several orders of magnitude higher than deduced here. This discrepancy can be understood by reduced response phenomena and thus smaller differences between He^+-metal and He^0-metal interaction potentials close to the surface [16]. This is scenario (2) proposed by More *et al.* [14] and by van Someren *et al.* [15].

In conclusion, we have observed fractions of He^+ ions which survived grazing scattering from a Ag(111) surface. This survival of ions provides clear evidence for neutralization of ions in the vicinity of the distance of closest approach to the surface plane, i.e., close to the jellium edge and image plane. As a consequence, Auger neutralization rates are found to be some orders of magnitude smaller than derived in many previous studies on neutralization of noble gas ions at metal surfaces. Recently, refined calculations on Auger transition rates and analysis of experiments reported results as confirmed by our work. In this respect we point out that the observation of fractions of survived ions provides the most direct information on the interaction scenario achieved so far for Auger neutralization [preliminary data obtained with a Cu(111) target show similar effects]. Finally we mention that the neutralization process might be affected by the projected *L*-band gap of Ag(111) [6] so that studies for an Al target—the prototype of a free-electron metal—are of considerable interest for future studies on this problem.

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