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Atomic and fullerene ions interacting with metal surfaces

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Abstract. Recent progress of studies on the interaction of keV atomic and fullerene ions during grazing scattering from solid surfaces is reported. We focus on electron capture processes for two systems that have been investigated in detail during the last years, the neutralization of He and C$_{60}$ ions at metal surfaces. Both species can be considered as model systems for Auger neutralization and for resonant neutralization of a large molecule, respectively. Basic experimental concepts and results obtained for scattering from an Al(100) surface are discussed.

1. Introduction
During the last decades, a large body of work was devoted to studies on the interaction mechanisms during scattering of atoms, molecules, and their ions from solid surfaces [1–4]. A main motivation for these efforts are promising long-term objectives, such as a predictive microscopic understanding of chemical reactions at surfaces (heterogenous catalysis), surface analytical tools (e.g., SIMS, LEIS, or STM), or plasma-wall interactions. In this respect, charge transfer plays an important role. The transfer of electrons for atomic or molecular particles in front of solid surfaces is initiated as soon as the potential barrier is sufficiently lowered. In front of metal surfaces, two mechanisms dominate: (1) resonant electron transfer, an elastic tunneling process where the energy of the transferred electron is conserved, and (2) Auger capture, an inelastic tunneling mechanism, where the total energy is conserved via excitation of the solid (e.g., electron-hole pair or plasmon) mediated by Coulomb interaction.

Here, we focus on experimental studies on the interaction of keV atoms and molecules scattered under grazing angles of incidence of about $\Phi_n \sim 2^\circ$ along high-indexed directions from surfaces. Scattering proceeds in the regime of surface channeling, characterized by a series of small angle scattering events with negligible energy transfer to the surface and effective decoupling of the motion parallel and normal with respect to the surface [2, 5]. Whereas for projectiles with energies $E$ in the keV range the motion parallel to the surface proceeds with constant velocity or energy $E_{\parallel} = E \cos^2 \Phi_n \approx E$, the effective interaction energy (normal energy) with the surface $E_{\perp} = E \sin^2 \Phi_n$ is of the order of eV. The fast parallel motion provides an efficient detection of scattered projectiles, whereas the slow normal motion allows one to probe charge transfer rates and energy level shifts in the domain of a few a.u. (atomic units are used unless otherwise stated) in front of the topmost layer.

In the following, we discuss two diverse systems that have gained attention in recent years, namely the interaction of He (light noble gas atom) and C$_{60}$ (prototypical large molecule) ions with metal surfaces. We focus on electron capture processes. The basic concepts are outlined for an Al(100) surface (prototype of a free electron metal). Figure 1 shows a sketch for the interaction scenario of a
He\(^+\) ion (left panel) and a C\(_{60}\)\(^{2+}\) ion (right panel) as function of distance from an Al surface (middle). The fullerene ion comprises of a quasi-continuum of unoccupied electronic states degenerate with the occupied part of the Al conduction band resulting in an efficient resonant electron capture (RC). The shifted unoccupied energy levels of He\(^+\) in front of the surface do not allow for RC, so that the dominant neutralization channel for He\(^+\) ions in front of high work function (\(W > 4\) eV) metal surfaces is Auger capture / Auger neutralization (AC / AN) into the ground state [3]. For both systems, we could recently determine the effective distances of electron capture.

Figure 1. Sketch of interaction scenario for electron capture by He\(^+\) (left) and C\(_{60}\)\(^{2+}\) (right) in front of Al surface (middle).

2. Interaction of He ions during grazing scattering from Al(100)

Until recently, the understanding of the neutralization of He\(^+\) ions approaching metal surfaces with eV energies was dominated by a pronounced discrepancy between experimentally [6–8] and theoretically [3, 9, 10] determined neutralization distances \(z_{\text{AN}}\). The experimental estimates were derived from measuring the He 1s levelshift \(\Delta E_{1s}(z_{\text{AN}}) \approx 2\) eV at the distance of neutralization via shifts of the high energy cutoff of Auger electron energy spectra [6, 7] or from the normal energy gain \(E_{\text{gain}}(z_{\text{AN}}) = V_{\text{He}0}(z_{\text{AN}}) - V_{\text{He}+}(z_{\text{AN}}) = \Delta E_{1s}(z_{\text{AN}})\) of He\(^+\) ions due to the change of the interaction potential \(V\) at the instant of neutralization [8]. Assuming a levelshift \(\Delta E_{1s}(z) = 1/4z\) based on the classical image charge interaction, experimental estimates consistently yielded \(z_{\text{AN}} \approx 6–7\) a.u. (reference: topmost surface layer) in shear contrast to theoretical predictions of \(z_{\text{AN}} \approx 3\) a.u. The corresponding experimental and theoretical AN rates differed by several orders of magnitude.

The levelshift turned out to be the key issue for the understanding of this discrepancy. Using an improved theoretical model, More et al. [11] predicted pronounced deviations from the classical behavior for distances \(z < 7\) a.u. Based on refined theoretical levelshifts, van Someren et al. [12] reanalyzed experimental Auger electron energy distributions and shifts of angular distributions and found similar results for \(z_{\text{AN}} \approx 3\) a.u. as well as for \(z_{\text{AN}} \approx 6–7\) a.u., but were unable to experimentally distinguish between the two scenarios. Experimental evidence for a neutralization close to the surface was provided by the experimental finding that small fractions \(P^+ \approx 10^{-3}–10^{-4}\) of He\(^+\) ions, grazingly scattered from metal surfaces, survive the complete scattering event in their initial charge state [13, 14]. For \(z_{\text{AN}} \approx 6–7\) a.u., the AN rates would result in fractions about 100 orders of magnitude smaller. The picture was completed by studies of Auger electron distributions [15] and normal energy gains \(E_{\text{gain}}\) for grazing scattering of He\(^+\) ions [16] that provided the experimental proof that the levelshift is strongly reduced close to the surface down to negative values. These studies did clear up the discrepancy between experimentally and theoretically determined neutralization distances and provided also direct access to the AN rate and He 1s levelshift in the interesting region of some a.u. in front of the surface, close to jellium edge and image plane.

For normal energies in the eV range, the distance of closest approach during surface scattering can be shifted on a scale of a.u. Therefore, fractions of surviving ions \(P^+(E/\text{eV})\) and normal energy gains...
\( E_{\text{gain}}(E_{\text{in}}) \) as function of incident normal energy \( E_{\text{in}} \) provide information on the distance dependent AN rate \( \Gamma_{\text{AN}}(z) \) and He 1\( s \) levelshift \( \Delta E_{1s}(z) \) via integration along different trajectories,

\[
P^+ = \exp\left(-\int_{\text{traj.}} \Gamma_{\text{AN}}(z) \, dz\right) \quad \text{and} \quad E_{\text{gain}} = \frac{\int_0^E E_0 \, dP^+}{\int_0^E dP^+} .
\]

This tomographic method has been applied to the study of AN rates and levelshifts for He in front of a variety of metal surfaces [13, 14, 16–21, 24]. In most cases, a detailed microscopic understanding of the interaction scenario could be achieved. Here, we demonstrate the experimental concepts for the interaction of \(^4\)He with an Al(100) surface [21].

In our experiments, \(^4\)He\(^0\) atoms and \(^4\)He\(^+\) ions with energies \( E \) of some keV are scattered under a grazing angle of incidence \( \theta_{\text{in}} \) from an atomically clean and flat Al(100) surface. The charge states for scattered projectiles are dispersed in an electric field, so that angular distributions for projectiles in different charge states can be measured simultaneously with a position-sensitive microchannel plate (MCP) detector. For details concerning the preparation of the surface and the experimental setup, we refer to [21]. In the upper panel of Figure 2, we show typical raw data for the measurement of charge state distributions (fractions of surviving ions). The \( y \) (z) direction corresponds to the azimuthal (polar) scattering angle. Two charges (He\(^0\) and He\(^+\)) with about 3 to 4 orders of magnitude different intensities are identified. Due to the background of the MCP of about \( 10^{-3} \), the two charges are measured separately, as indicated by the vertical line. In the lower panel, we show measured ion fractions as function of normal energy \( E_{\text{norm}} \) for incident \(^4\)He\(^+\) (full symbols) and \(^4\)He\(^0\) (open symbols, upper limits) with energies \( E = 2 \text{ keV (squares)} \) and \( E = 3 \text{ keV (circles)} \). For incident ions, the ion fractions are about two orders of magnitude larger than for incident atoms. Therefore, the ion fractions in the scattered beams for incident ions are explained by an incomplete neutralization, whereas reionization of neutralized projectiles does only contribute up to about 1% to the ion fractions and can be neglected.

The data is compared to 3D Monte Carlo simulations based on theoretical AN rates and levelshifts. For details see [21]. The full triangles (connected by curve to guide the eye) show results based on an AN rate calculated using a LCAO basis set for the on top position in the lateral unit cell [22] which are about a factor five larger than the experimental data. Due to the extreme sensitivity of the survival probability \( P^+ \) on the AN rate \( \Gamma_{\text{AN}} \) (cf. Equation (1)) the difference corresponds to an increase of the
latter by a factor 1.27 (full diamonds with curve), only. Therefore, the agreement is on the level of the intrinsic uncertainties of DFT calculations of AN rates of about 20% due to approximations, such as neglect of indistinguishability of the transferred electron from other electrons at the surface [9, 10, 21].

![Figure 3](image)

In Figure 3(a) and (b), we show polar angular distributions for outgoing He\(^0\) for incident He\(^+\) (curves with full symbols) and He\(^0\) (curves with open symbols) with (a) \(E = 2\) keV and \(E_{\text{L}m}\approx 1\) eV and (b) \(E = 15\) keV and \(E_{\text{L}m}\approx 19\) eV. For the small incident normal energy, the distribution for neutralized ions is shifted to larger polar exit angles \(\Phi_{\text{out}}\) (with respect specularly reflected He atoms) corresponding to a larger outgoing normal energy \(E_{\text{L}m}^{\text{out}} = E \sin^2 \Phi_{\text{out}}\) a positive normal energy gain \(E_{\text{gain}} = E_{\text{L}m}^{\text{out}} - E_{\text{L}m}^{\text{in}}\), and a positive He 1s levelshift of about \(\Delta E_{1s(z_{\text{AN}})} \approx 2\) eV at the effective distance of neutralization. For the larger normal energy (panel (b)), \(z_{\text{AN}}\) is located closer to the surface. At this distance, the relative outgoing angles of the angular distributions are reversed and give a negative normal energy gain or levelshift at the distance of neutralization of \(\Delta E_{1s(z_{\text{AN}})} \approx -1\) eV. Due to the larger \(E_{\text{L}m}\), the relative effects are reduced, but can still be detected within the experimental angular resolution of about 0.01°. The He 1s levelshift changes sign close to the surface.

Panel (c) shows the normal energy gain \(E_{\text{gain}}\) as function of the incident normal energy \(E_{\text{L}m}\) (full circles) obtained from angular distributions as shown in Figures 3 (a) and (b). As for the fractions of surviving ions, the data is in good accord with the 3D Monte-Carlo simulations (open circles) [21, 22]. We thus state a detailed microscopic understanding of the interaction of He\(^+\) ions and He\(^0\) atoms with an Al(100) surface.

Based on these concepts, a number of studies have been published in recent years comparing the interaction for monocrystalline surfaces of different orientation [17, 18, 20, 21], discussing isotope effects [18, 21], and analyzing the role of the electronic structure (\(d\)-electrons, surface state, spin-polarization) on the neutralization rate as well as its lateral corrugation [19, 20, 23, 24]. Furthermore, the microscopic understanding of the neutralization regime at low energies \(E \leq 3\) keV has been extended to the region of intermediate energies \(E = 3–10\) keV where additional charge transfer mechanisms, in particular ionization, play a role [25, 26].

3. Interaction of C\(_{60}\) ions during grazing scattering from Al(100)

Compared to the large body of literature on the interaction of hyperthermal atomic projectiles with solid surfaces, studies with larger molecules or clusters are rare and suffer from an enhanced complexity introduced by the internal degrees of freedom of the molecules [1–4, 27, 28]. The symmetrical shape, well-known electronic structure, simple production, and the large variety of
applications [29] of fullerenes make \( C_{60} \) a promising candidate for exploring molecule/cluster-surface interactions. The majority of \( C_{60} \) surface scattering studies have been performed under large impact angles, where the outcome of the experiments is affected by pronounced energy transfers to and deformations of the surface [27, 28, 30–34]. We mention that until recently, besides the observation that positively charged \( C_{60} \) ions are efficiently neutralized at metal surfaces, no detailed experimental information on the distances of electron capture was available. First experiments in the grazing geometry with a negligible energy transfer to the surface were reported by Kimura et al. for a KCl(100) surface [35, 36]. Here, we summarize recent experiments for grazing scattering of positively charged \( C_{60} \) molecules from an Al(100) surface [37–39].

In our experiments we have scattered \( C_{60}^+ \) and \( C_{60}^{2+} \) ions under grazing angles of incidence from a well-prepared Al(100) surface. These beams were produced by evaporation of \( C_{60} \) powder in an ECR ion source. The experimental setup is the same as mentioned above and we refer to [38] for details. We did not succeed to generate defined beams of neutral molecules.

For atomic projectiles in front of metal surfaces, resonant charge transfer (if possible) proceeds relatively far from the surface (compared to AN), where the levelshift and the interaction potentials can be approximated by the concept of classical image charges [1–4]. We expect a similar case for electron capture by positively charged \( C_{60}^{2+} \) ions in front of a metal surface (cf. Figure 1) and study the distance for electron capture for a \( C_{60}^{2+} \) ion indirectly via energy gains deduced from shifts of angular distributions. Owing to the lower potential barrier for a doubly charged projectile, the neutralization of doubly charged clusters is expected to occur in two well-separated electron capture events, so that the difference in normal energy gain for doubly and singly charged ions is given by

\[
\Delta E_{\text{gain}} = V_+ (z_{2+ \rightarrow +}) - V_{2+} (z_{2+ \rightarrow +}),
\]

where \( V_+ \) (\( V_{2+} \)) is the interaction potential for the singly (doubly) charged cluster and \( z_{2+ \rightarrow +} \) is the distance of the first electron capture to \( C_{60}^{2+} \). The additional energy gain of the doubly charged cluster \( \Delta E_{\text{gain}} \) can be compensated by an increase of the incident normal energy of the singly charged cluster by the same amount (\( \Delta E_{\text{gain}} \)) resulting in the same outgoing normal energy for doubly and singly charged clusters.

![Figure 4](image_url)

**Figure 4.** Panel (a): outgoing normal energy as function of incident normal energy for scattering of \( C_{60}^+ \) (full circles) and \( C_{60}^{2+} \) (open circles) from Al(100). Open squares (open triangles): incident normal energy of \( C_{60}^{2+} \) data shifted by 1.41 eV (1.60 eV). Panel (b): difference in normal energy gains of \( C_{60}^{2+} \) and \( C_{60}^+ \) scattered from Al(100). Open circles: experimental data. Dotted (dashed) line: prediction of COB model for pointlike projectile (conducting sphere). For details see text.

In Figure 4 (a), we show outgoing normal energies as function of incident normal energy derived from the maxima of angular distributions for incident \( C_{60}^{2+} \) (open circles) and \( C_{60}^+ \) (full circles). In addition, we show data for \( C_{60}^{2+} \) for a shift of the incident energy scale by \( \Delta E_{\text{gain}} = 1.41 \) eV (open squares) and \( \Delta E_{\text{gain}} = 1.60 \) eV (open triangles), corresponding to the prediction of a classical over-
barrier (COB) charge transfer model, where the fullerene is described by a conducting sphere [40] and by a pointlike (pseudo-atomic) projectile, respectively [37]. A direct comparison of the experimental difference of normal energy gain $\Delta E_{\text{gain}}$ with the theoretical predictions of the two COB models is presented in panel (b). The data is in good accord with the COB model for a conducting sphere (dashed line), whereas the agreement with the COB model for the pointlike projectile (dotted line) turns out to be poorer. The main difference between the two models is the polarizability of the sphere which obviously needs to be taken into account. The COB model for the conducting sphere yields the first information on a distance for electron capture to $C_{60}^{2+}$ of 14.8 a.u. [37], where the closest C atom of the fullerene is slightly more than a fullerene cage radius in front of the surface.

Figure 5. Outgoing normal energy as function of incident normal energy for scattering of \( C_{60}^{+} \) ions from Al(100) (circles) [38, 39] and KCl(100) (squares, data by Matsushita et al. [36]) derived from the positions of the maxima (full symbols) as well as the positions of the half-maxima (open symbols) of polar angular distributions are given in Figure 5. Whereas the majority of projectiles is scattered with a maximum outgoing normal energy of about 7 eV for the metal surface, the clusters are scattered elastically for normal energies up to 20 eV in case of the insulator surface. The latter behaviour is in accord with classical 3D trajectory simulations for the KCl(100) (downward triangles with curves) as well as the Al(100) surface (upward triangles with curves) based on the empirical Tersoff potential [41] for the cluster (for details see [38, 39]). The differences for the Al(100) surfaces can not be attributed to the interaction potential with the surface, as simulations for a wide range of interaction potentials yield similar results. They also can not be attributed to deficiencies of the Tersoff potential which describes the data for KCl(100) fairly well. Therefore, the inelastic scattering for the Al(100) surface is most likely due to a perturbation of the electronic structure of the fullerene by the metal surface which affects the elastic properties via a coupling of electronic and phononic degrees of freedom.

For the Al(100) surface, the normal energy loss is completely transferred to internal degrees of freedom of the fullerene as demonstrated by the analysis of fragment spectra for scattered clusters as shown in Figure 6 (a) [38, 39]. Based on the concept of microcanonical temperature [42] and an Arrhenius representation of the rates for delayed electron emission and $C_{2}$ emission [43], the formation of charged fragments during the flight time of several $\mu$s can be calculated as function of internal energy (cf. panel (b)). From a fit of the fragment spectra for scattered clusters, the internal energy distribution can be determined [38].
4. Summary and conclusions

In this paper, we have outlined recent progress in studies on the neutralization of He and C$_{60}$ ions. The substantial differences in electronic structure and arrangement of nuclei result in fairly different interaction regimes during scattering from the surface. Our studies were performed in the regime of scattering under grazing angles of incidence (“surface channeling”) where the impact of fast projectiles proceeds in the hyperthermal energy regime, i.e. kinetic energies for the motion normal to the surface plane in the eV range. The high ionization potential of the He atoms gives rise to the dominance of the Auger process for the neutralization of He$^+$ ions. Key issue of the studies was the observation of very small but well defined fractions of ions which survive the complete scattering event and open the way for the application of methods of tomography for the analysis of data in terms of distance dependent Auger neutralization rates. Furthermore, for surface channeling, trajectories of atoms and ions are affected by dielectric response phenomena which are closely related to electronic level shifts of the interacting species. Combined application of these techniques provides clear-cut and new information on the AN process where the pronounced deviation of the level shift from the monotonic increase for reduced distances has led to a revised interpretation of former experiments. Based on this approach, AN takes place much closer to the surface plane so that the discrepancy between experimentally derived and theoretical AN rates could be resolved. In fact, within in the limits of accuracy of current theoretical ab-initio methods one can state quantitative understanding for this electron transfer process.

For C$_{60}$ ions, charge transfer proceeds via resonant electron tunnelling where the extended geometrical structure of the fullerene affects the tunnelling potential barrier and charge exchange. By making use of image charge effects on the trajectories of scattered ions, we could deduce the effective distance of resonant electron capture to C$_{60}^{2+}$ ions. Similar as shown in previous studies with an insulator surface, we observed that keV fullerenes are scattered specularly as long as the impact energy for the motion along the surface normal does not exceed some eV. The normal energies for the pronounced sub-specular scattering observed for metal surfaces at larger energies are clearly smaller than for insulator targets. The origin for this effect is presently not cleared up and under active research.

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