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Highly charged ions interacting with thin-film covered surfaces

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One of the most appealing properties of highly charged ions is their high potential energy, *i.e.* the sum of their ionization potentials. This energy has to be provided during ion production, and in turn it is available when these ions come into contact with neutral matter. For $\operatorname{Ar}^{10+} e.g.$ a total energy of about 10 keV is available. Since a long time it is discussed whether this energy can be exploited *e.g.* to induce surface modifications with *nm* dimensions. For sufficiently low kinetic energy of the ions this would be possible without bringing any damage into deeper layers of the target.

Alternatively highly charged ions can possibly be used as "probes", *e.g.* to investigate magnetic ordering of surfaces or thin films. Here we want to use the fact that many electrons are simultaneously captured from a very small surface area. High-spin states of the neutralized projectile are expected to result from such capture processes when the captured electrons stem from a ferromagnetically ordered area. Spectroscopy of these states should therefore enable the detection of magnetic ordering. Compared with other techniques this method is expected to be extremely surface sensitive and can moreover detect short-range ordering on a nm scale!

In the last years we have studied various processes which can be regarded as basis for such applications and therefore should be understood beforehand. We have used Auger electron spectroscopy to study the electronic processes taking place near clean and thin-film covered surfaces upon highly charged ion approach.^{1–3)}

Hollow atom formation near clean metal surfaces

Electron capture into excited orbitals of highly charged ions gives rise to the formation of so-called hollow atoms, in particular near metal surfaces. Since these hollow atoms decay via Auger processes with typical lifetimes of fs, electron spectroscopy is an appropriate method to obtain information on the various processes taking place near the surface. A typical electron energy spectrum resulting from collisions of hydrogenlike N⁶⁺ on Ni(110) is shown in Fig. 1. One can clearly distinguish the LMM and the KLL Auger electrons. A closer analysis of the KLL part of the spectrum reveals details of the electron dynamics near the surface. Especially the intensity ratio of the two peaks at the low- and the high-energy side of the KLL electron distribution respectively is a sensitive measure for the L-shell filling rate.

These peaks result from decay processes with the L-shell filled with two electrons (configuration $1s2s^23l^4$) or six electrons ($1s2s^22p^4$) respectively. For an L-shell filling rate that is slow as compared to the Auger rate the KLL process will take



Fig. 1. Energy spectrum of electrons resulting from N^{6+} + $\mathsf{Ni}(110)$ collisions.

place as soon as two electrons have arrived in the L-shell and the low-energy peak dominates. For increasing L-shell filling rate the high-energy peak becomes more and more important. In fact the sharp peak at the low energy side can be ascribed to above-surface processes on the incoming trajectories.

The fact that already in front of the surface an appreciable amount of the projectile's potential energy is converted into kinetic energy of emitted electrons implies that only part of that energy is available for possible surface modifications. Moreover metals are less suited for surface modifications because electronic response times are much too short.

Electronic processes near insulating surfaces

More promising candidates for surface modifications are insulator surfaces. We therefore have measured electron spectra resulting from highly charged ion collisions on insulating LiF surfaces and have found that indeed the above-surface component in these spectra was absent. However, when replacing a metal by an insulator surface, several parameters are changed simultaneously - e.g. conductivity, binding energies of the electrons, as well as band gap and band structure. The existence of the band gap can e.g. prohibit an effective exchange of electrons between the solid and the projectile, because there are no states available in the band gap into which electrons - promoted due to image charge effects - could be transferred.



Fig. 2. KLL part of electron spectra resulting from O⁷⁺ collisions on Au(111) and LiF(100) (left panel) and on an Au(111) surface covered with varying amounts of LiF (right panel).

To see which of the various parameters are mainly responsible for the modified electron dynamics we have employed thinfilm covered surfaces as targets, thereby changing only one of the parameters. Figure 2 shows electron spectra arising from O^{7+} collisions on an Au surface, covered with various fractions of a LiF monolayer. After coverage with one monolayer the electron spectrum resembles more or less that of a bulk LiF surface. This implies that the bandstructure of LiF, which is not yet developed in one monolayer, can not be responsible for the changes in the electron spectra.

We conclude from this that it is mainly the significantly increased binding energy of the electrons (12 eV instead of about 5 eV) which is responsible for the modification of the electron spectra and thus for the suppression of the above-surface processes.

Electron spectra from $C_{\scriptscriptstyle 60}$ covered Au and from carbon (HOPG) surfaces

In order to study in more detail the effect of the electronic structure of the surface we have measured electron energy spectra resulting from highly charged ion collisions on various carbon surfaces. Especially C₆₀ covered metal surfaces are interesting from several points of view. It might e.q. be possible to create nanostructures via the fragmentation of C_{60} present as a monolayer or a thin film on a suitable target. Whereas pure C_{60} is semiconducting, its conduction band is partly filled if it is formed as a monolayer on top of e.q. Au. In that case it will acquire metallic properties with the same workfunction as Au. However, the electronic surface density of states can be quite different from that of a pure Au surface. Figure 3 shows a comparison of KLL Auger spectra of electrons, emitted during collisions of 3 keV O^{7+} ions on a pure Au surface, an Au surface covered by a monolaver of C_{60} , and a pure carbon surface (highly oriented pyrolytic graphite). A remarkable feature is the fact that the low-energy peak is very pronounced for the clean Au(111) surface, whereas the high-energy peak is especially pronounced in case of the C_{60}

covered Au surface. This can be attributed to a slow filling of the projectile L-shell via Auger cascades in the hollow atom far in front of the clean Au surface. For the C₆₀ covered Au(111) surface on the other hand the filling of the L-shell is apparently starting at significantly smaller distances from the surface, but at an increased filling rate. Similar differences in the spectra are also seen for other collision energies and other hydrogen-like projectiles. At the moment we can only speculate that the fast L-shell filling in case of the C₆₀ covered Au(111) might be connected with an especially high electronic surface density of states.



Fig. 3. Energy Spectra of Auger electrons emitted during collisions of 3 keV O^{7+} ions on a clean Au(111) surface (top spectrum), an Au surface covered with one monolayer of C₆₀ (middle spectrum), and a HOPG surface (bottom spectrum).

Future experiments for the analysis of surface magnetism

As pointed out in the introduction highly charged ions might be used as probe for (short range) magnetic ordering with an extreme surface sensitivity. This is based on the fact that some peaks in the electron spectra can clearly be identified as being due to the formation of singlet and triplet states respectively. The two peaks in the lower left spectrum of Fig. 2 at 465 and 480 eV respectively are identified as being due to a $(2s^2)^{1}S$ and a $(2s2p)^{3}P$ L-shell formation respectively. If spins are conserved the $(2s^2)^{1}S$ peak should be strongly suppressed in case of electron capture from a ferromagnetically ordered surface. An interesting aspect of this method is the fact that short range ferromagnetic ordering can be detected even if the orientation of the spin ordering is changing, e.g.in neighboring domains or terraces: since electron capture by each specific ion takes place from an area with the spatial extension of nm the actual diameter of the ion beam is not limiting the "resolution" of this method.

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