Hollow-atom probing of surfaces

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Objectives

In the last two decades, the development of powerful ion sources has facilitated the use of relatively slow, intense beams of highly charged ions as a tool in physics research. Originally, such beams were used to study the interaction of highly charged ions with gaseous targets (see e.g. Janev and Winter). At present, such experiments are still carried out and results are used in other fields of physics, for instance in thermonuclear fusion research. However, it was soon recognized that highly charged ions could also be used as tools in fundamental and applied surface physics. The use of ion beams to probe solid surfaces has a long tradition which goes back to the early fifties when Hagstrum carried out numerous ion-beam experiments to study ion-surface interactions. The interaction of ions with solids can in general be separated in kinetic processes (as ion scattering and sputtering) and electronic processes (ion neutralization and deexcitation). For highly charged ions, the electronic processes are of particular interest in view of the large amount of potential energy carried by these species. The potential energy carried by highly charged ions (equal to the sum of the successive ionization energies) is comparable to – or may even exceed – the kinetic energy of these species. If such a highly charged ion collides with a solid surface intriguing situations arise in which many electrons are captured by the ion even before the surface is reached. The reaction products of such a collision may reveal detailed information, not only on the processes leading to the energy release of the ion but also on the electronic structure of the target surface. Interest in the field of highly charged ion-surface interactions is however not only driven by the opportunity to study novel atomic or
solid state phenomena on a fundamental level. Also the promise of future applications of highly charged ions as a tool to create surface structure on a nanometer scale has been an important motivation for research.

At the KVI, Siebe de Zwart started investigating the interaction of multiply charged ions with surfaces in 1983. At that time the KVI had just started using an “electron cyclotron resonance ion source” to produce beams of highly charged ions. De Zwart and coworkers designed and built the present surface physics set-up nick-named Sirè. This set-up has the rather special property that the highly charged ions can be decelerated down to less than one thirtieth of their initial velocity (of about 1000 km/s). Such slow ions spend a relatively long time in front of the target surface mounted inside the setup. This way, it is assured that the processes leading to neutralization and deexcitation of the projectiles can be observed with great detail. By studying the electrons emitted during collisions of slow, multiply charged Ar ions on a Si surface, De Zwart found out that these ions reached a significant degree of deexcitation already before they actually hit the surface. In another series of experiments, in which he studied the yield of sputtered (Si) target material as a function of the charge of the impinging ions, he found however no evidence for a so-called Coulomb explosion (see the preface) predicted by Bitenskii and coworkers.3

In 1987, Luuk Folkerts continued the work of De Zwart. Folkerts investigated the energies and emission angles of electrons emitted during collisions of highly charged carbon, nitrogen, oxygen, and neon ions on tungsten and nickel surfaces. By varying – among other things – the charge and velocity of the incoming ions, he was able to distinguish between electrons emitted from projectiles still above the surface and from projectiles which had already entered the surface. At that time it became clear that the energy carried by highly charged ions is dissipated in a rather small volume starting a few nanometers in front of the surface. Just before Folkerts finished his PhD work, Burgdörfer and his coworkers4 presented a theoretical model by which one could actually calculate this starting point. In this model, the ion’s energy release starts more or less at the same time as a “hollow atom” – a highly excited but neutral species with many electrons in loosely bound outer shells – is formed by resonant capture of valence band electrons. However, the course of this energy release – i.e. the evolution of the hollow atom created in front of a surface – was at that time to a large extent still unknown.

Those results were, very briefly, the premises of the research leading to this thesis. The questions which will be addressed in this work can be summarized as follows:
I. What are the temporal and spatial dimensions of the energy dissipation of highly charged ions interacting with solid surfaces?

II. What is the nature of the electron exchange between ion and solid leading to this energy dissipation?

III. Do the reaction products of highly charged ion-surface collisions facilitate the use of such ions as tool to probe surface structure?

These questions are addressed using experiments which study the reaction products resulting from collisions of hydrogenic ions with metal, semiconductor, and insulator surfaces. A hydrogenic ion is almost completely stripped except for one remaining electron which resides in its K-shell. The ions that we used all were of first-row elements.

The products of the ion-surface collisions can be grouped into (Auger) electrons, scattered projectiles and sputtered target material. The work in this thesis concentrates primarily on (i) KLL Auger electron spectra, and (ii) charge state distributions and energy loss of scattered projectiles.

Ad (i): The deexcitation of hollow atoms formed in front of a surface is accompanied by the emission of many electrons. One of the most important processes leading to potential emission of electron is the Auger transition. Classically, an Auger transition can be considered as a collision of two electrons, by which one electron is transferred into a deeper shell and the other one is emitted into the vacuum. This electron, the “Auger electron” can be measured. The label ‘KLL’ implies a transition in which one L-shell electron is transferred to the K-shell and another L-electron is emitted.

Ad (ii): For certain circumstances, projectiles impinging on solid surfaces are scattered off the surface. The charge state distribution of these projectiles and the energy lost during the collision with the surface yields detailed information on the processes playing a role during the ion-surface interaction.

The plan of this thesis

In short, the plan of this thesis is as follows:

- The next chapter discusses the theoretical concepts used to describe highly charged ion-surface interactions. An overview of the experimental techniques will also be given in this chapter.
A highly charged ion approaches a surface. Somewhere in front of the surface, a hollow atom may be formed. Inside the solid, such a highly excited particle cannot exist. The question mark reflects the objectives listed in the text.

- Chapter 3 presents a discussion of the experimental techniques used. The ion source and Sirφ experimental set-up will be discussed.

- In chapter 4 the basics of Auger electron spectroscopy will be introduced using KLL Auger spectra obtained from highly charged ions impinging on different metals and on silicon. Spectral features are discussed taking the electronic structure of the different targets into account.

- Chapter 5 presents a theoretical discussion of the shape of KLL Auger spectra obtained from ion-surface collisions. The model developed will be applied in chapter 6 on spectra obtained from hydrogenic and metastable helium-like carbon, nitrogen and oxygen colliding with a silicon surface.

- In chapter 7, the model is extended to cover the different mechanisms governing the filling of the inner shell holes of the hollow atom. From spectra obtained from hydrogenic nitrogen ions on an aluminum surface, the trajectory along which the inner shell vacancies are filled (i.e. the volume in which the energy of the hollow atom is dissipated) is located.
• Chapter 8 presents a comparison between spectra obtained from hydrogenic ions impinging on conducting surfaces and on an insulating LiF surface. Differences between these spectra suggest the absence of hollow atom formation in front of the insulator for certain circumstances.

• In chapters 9 and 10, the charge states and energy loss of scattered projectiles are discussed. In chapter 9, the energy lost by He projectiles scattered off an aluminum surface is modeled. This is done in terms of the friction the projectiles suffer during their path through the ‘electron sea’ at an Al surface. In chapter 10 the charge state distribution and energy loss of oxygen ions scattered off the same Al surface are investigated for different incoming charge states.

• In chapter 11, the formation of negative ions is discussed. In these experiments singly- and multiply charged carbon, oxygen and fluorine ions are incident on an Al surface. When scattered off the Al surface, these projectiles can be ionized and thus form a negative ion. The processes leading to the formation of such a negative ion are investigated. The probability of negative ion formation shows a strong dependence on the orientation of the electronic orbitals of the projectile with respect to the surface.

Finally, chapter 12 evaluates the work presented in this thesis and addresses the objectives listed above.