Hollow Atoms Above Dielectrics And Metals

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Slow highly charged ions approaching surfaces at very close distances (~nm) are known to capture many electrons in highly excited states forming hollow atoms(1)(2). These hollow atoms quickly decay to their ground states through a long cascade of autoionization processes(Auger transitions) which may be balanced , as these ions are still close to the surface, by a series of electron captures. This actual sequence of many events of capture and autoionization, alternate or not, is not experimentally known to date, and depends on the charge of the ions and of the capture processes which may be very different above metals and insulators. We review in this paper two new experiments on the kinematics of fully decelerated highly charged ions(Ar^{18+}) above dielectric and metal surfaces, and on the behavior of ions of lower charge states (O^{7+} and Ne^{9+}) above the same surfaces.

Introduction

The behavior of slow highly charged ions above surfaces may be studied on line by looking at the Auger electrons and the X rays emitted during the decay of the ions to the ground state. The hollow atoms formed above the surfaces decay, at the beginning, when the electrons are in the outermost shells, by a cascade of Auger transitions, and later on, in the innermost shells, also by the emission of X rays. The filling of the innermost shells of the ions is, as experimentally demonstrated, a stepwise process(2). The last sequence of the cascade is the filling of the 8 holes of the L shell. In the considered experiments we used Hlike ions i.e. ions owning a single K vacancy, and looked at the K X ray emitted during the filling this K hole. The energy of this K line linearly depends on the number of L spectator electrons present at the time of the decay(satellites lines). These satellites may be separated in high resolution X ray spectroscopy. When this K line is emitted at any time of the filling of the L shell i.e. in presence of any number x of L spectator electrons, one observes statistically a characteristic array of 8 KL^x satellites as shown in Fig: 1. The intensities of these lines sign the relative rate at which the L and K shells are filled. When the filling rate of the L shell is slow, compared to that of the K hole, this distribution peaks to the lines corresponding to a small number of L spectator electrons (KL^1): the K X ray is emitted as soon as a first electron fills the L shell. In the opposite case(fast L shell filling) the distribution strongly peaks on the KL^8 line: the K line is emitted when the L shell is quasi closed. The rate at which the L shell is filled obviously depends on the initial process of capture. This so called 'atomic clock 'property of the hollow atoms has been extensively used in the last decade to study the behavior of these ions above and below the surfaces.

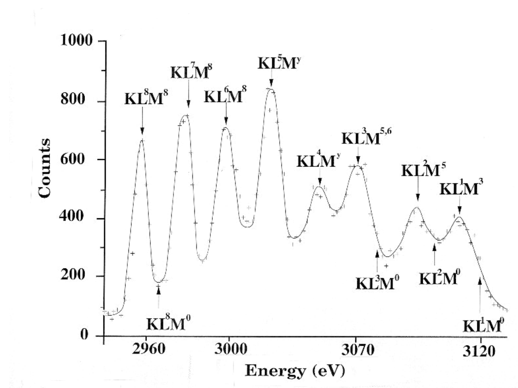

Fig:1. KL^x array of Ar satellites observed when the filling of the K hole holds at any time of that of the L shell

The main difficulty to observe the decay of the hollow atoms above the surfaces is the very short time T allowed to the ion to reach the surface from the distance z_0 at which it starts capturing electrons (few nm), compared to the meantime τ for the ion to fully decay to its ground state. In order to have enough time to observe these emissions before the ions hit the surfaces, they must be decelerated at the lowest possible velocities, to increase the travel time T. The deceleration of the ions is however limited by their image acceleration towards the surface. One cannot then decelerate the ions at energies lower than that of the image energy gain. This image acceleration leads to two consequences:

1) The so called saturation effect(3): at the lowest *given* ion kinetic energies the travel time T is only governed by this acceleration effect. If below z_0 the

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ion stays quasi neutralized, T is roughly constant (T ≤ 200 fs) when one decelerates the ion close to zero energy, and one does not observe at these lowest *given* kinetic energies any change of e.g. γ (number of ejected electrons) or of the spectral features.

2) The ion always touching the surface where it suddenly captures electrons in lower excited states (leading to a well known, but very different X ray spectrum), the observed X ray spectrum is then a blend of the above the surface decay under study, and the unwanted characteristic spectrum emitted below or at the surface.

The behavior of the hollow atoms above the surface has been studied by few groups in Auger spectroscopy for light ions such as O^{7+} or N^{6+}(4)(5) or Ar ions(6), and in X ray spectroscopy for the heaviest elements such as Ar^{17+}(1)(2).It is the purpose of this paper to compare in X ray spectroscopy, the behavior of the Ar^{17+} ions, above dielectric to that of ions of lower charge state(Hlike O and Ne).

Two main experiments have already been completed for Ar^{17+} ions.

1) Owing to the fact that one cannot a priori get away from touching the surface, and eliminates of the X ray spectra coming from the contact, one first studied the interaction of the considered ions with C_{60} targets which constitute some small surfaces. As the distance of interaction z_0 is much larger than the radius of the buckey balls, most of the ions approaching the fullerene targets may capture electrons at large distances, form hollow atoms, and then escape the capture area, not touching the surface. In this experiment(7) it was shown that an Ar^{18+} (bare) ion captures at close distances of the target many electrons in very high Rydberg states and decays later outside the capture area: the ion, not being re-fed in electrons, freely fully autoionizes, losing all its electrons but one to become, as observed, an Ar^{17+} ion.

2) In the second series of experiments Ar^{17+} (Hlike) ions were sent at normal incidence on metal and dielectric targets where they were fully decelerated, and we studied the evolution of the relative intensity of the first few satellites lines ($KL^{1,2,3}$), versus the ion kinetic energy, in the range of 0-12 eV/q i.e. of the order of magnitude of the image energy gains. These relative intensities were found, as expected(saturation effect), to be constant for metals and mainly displaying the characteristic X ray spectrum of the contact of the ion. Above dielectrics one observes, by contrast, a continuous change of these relative intensities. The observed spectra display the evolution, at decreasing ion kinetic energies, from that of the ion touch down, to that of the 'outside decay (the KL^1 line). A close inspection of this KL^1 satellite showed that this line was that of pure Helike ions (Ar^{16+*}) i.e. without any outermost spectator electron(8), like in the C_{60} experiment. The first finding(the vanishing of the saturation effect) was explained as due to the positive charges transient building up following the intense exodus of electrons during the approach of the ion of the insulator surface, which easily overbalances the image acceleration(9), and then backscatters the ion. The second result (apparition of pure Helike ions at the lowest energies) was explained(8) as due to the sudden decrease of the distance of first capture z_0, when the ion is strongly neutralized and the surface charged(z_0 varies as the inverse of the binding energy and the square root of the actual charge of the ion): the ion quickly captures many electrons and escapes the capture area, allowing a free autoionization of the hollow atom(no refeeding), like in the C_{60} experiment. We present in the next § a direct proof of the backscattering of the ions on dielectrics, and of this sequence of successive waves of quasi full neutralizations followed by full autoionization processes.

1) The kinematics of Ar hollow atoms above dielectrics

The kinematics of the hollow atoms above the surfaces has been studied at the lowest velocities(by biasing the target in a special device), with Ar^{18+} ions on Si targets. These ions which own two K vacancies will then display two times the atomic clock property. We used for this experiment three SiLi detectors(1,2,3) in coincidence, two on both sides of the target (detectors 1 and 2) and one 25 cm upstream in front of a pinhole(detector 3) where the incoming beam is passing through (Fig.2) . At energies larger than 12eV/q we observed with the two detectors in front of the target the typical coincidence spectra between the hypersatellite and satellite i.e. the filling of the first K hole (hypersatellite), followed by the filling of the second one(satellite). Both K

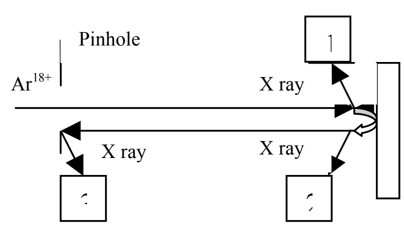

Fig:2. Backscattering of the ions

holes are then filled above the target. At lower energies this coincidence vanishes and one observes delayed coincidences between one of the detectors in front of the target(observation of the hypersatellite line: filling of the first K hole) and that located upstream (detector 3): observation of the satellite filling of the second K hole. The delay time between the emission of the K X ray above the target and that upstream has been found(fig.3) equal to the time of flight between detector 1 and 3.

These results mean that the first K hole of a same ion is emitted above the target, and the second one, on its way back while touching the edges of the hole due to imperfect alignment of the beam. Owing to the difficulty of perfectly controlling the direction of the ion beam through a one mm hole one deduced from the measurement of the coincidence rate that at least 70% of the incoming ions, but very likely much more, experienced this backscattering. These results, in good agreement with all previous spectroscopy experiments, mean that like in the case of the interaction of these ions with C_{60} targets the ion captures many electrons

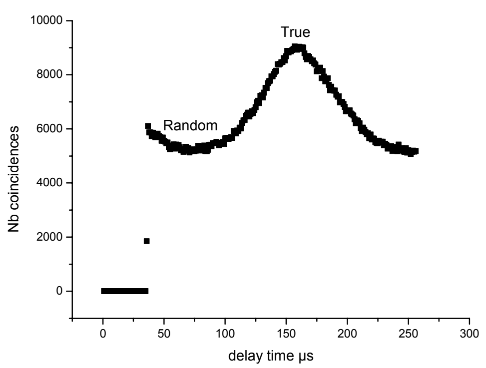

Fig:3. Number of coincidences versus the delay time between the detection in detector 3 with respect to 2.

above the target and, not being re-fed, owing to the quick decrease of the maximum distance of capture, freely autoionizes losing all its electrons but one. This now Ar^{17+} ion is then re-accelerated toward the hole, hitting one of its edges at the ground potential, displaying the well known characteristic satellite spectrum observed at full energy. The aim of the next section is to compare the behavior of these ions with that of ions of lower charge states.

2)The experiments on O^{7+} and Ne^{9+} ions

The same kind of spectroscopy experiments as those already done for Ar^{17+} ions have been carried out for Hlike O and Ne ions. In order to get a good compromise between the resolution of the detectors and their transmission we used SiLi detectors and Superconducting Tunnel Junctions (STJ) of 20 eV resolution. We present in Fig:4 the X ray spectra of O ions on Si observed with an STJ detector(10) for ions of 10 eV/q and nearly zero kinetic energies, which we compare to that observed at 10keV/q energy, where the ion emits its X ray below the surface while being fully neutralized. By contrast to what happens for Ar, where the KL^x distribution strongly peaks on KL^1, one observes for O ions two unresolved lines corresponding to all states of L ionization, in good agreement with previous investigations in Auger spectroscopy for similar ions(4)(5). The first line corresponds to the three unresolved $KL^{4,5,6}$ satellites, whose energy separations are lower than their natural widths or fine structures. The second broad line, centred on KL^2, is made of the three $KL^{1,2,3}$ satellites, equidistant in energy and more or less resolved. The fluorescence yield of the $KL^{4,5,6}$ states is of the

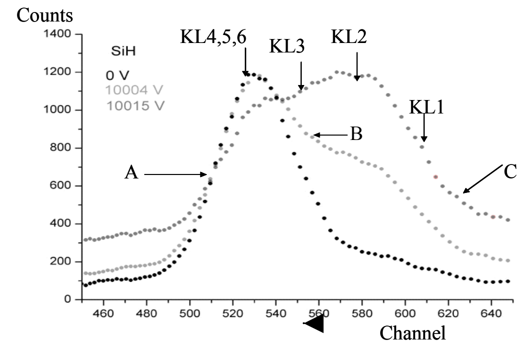

Fig:4. KL^x array of satellites observed with O^{7+} ions at various kinetic energies of the ion:. A: 10 keV/q, B: 10eV/q ,C: 2 eV/q.

order of 8 10^{-3}, while that of the $KL^{1,2,3}$ states is close to unity. If the second line is that of $KL^{1,2,3}$ states not owning any outermost shell electron, its relative intensity would be two orders of magnitude lower than it appears, and then negligible. As soon as these $KL^{1,2,3}$ states own outermost shell electrons, opening very fast Auger channels, their actual intensities would then be much larger. The conclusion is then that most of the observed states correspond to ions with a large number of L electrons (quasi neutralized), and of states with less L electrons, but few outermost shell electrons and then also quasi neutralized. These considerations based on the fluorescence yields allow then to state that part of the spectra comes from hollow atoms ($KL^{1,2,3}$ states).

Electron capture for O^{7+} holds in the n = 7 quantum states of the ions(11). The lifetimes of e.g. the KQ^{2-6} and KM^{2-5} states lie between.4 and 5 fs (12), and do not vary very much for the neighboring upper and lower states. It must not then take roughly more than about few fs or tens fs for the ion to complete the L shell filling, to be compared to a travel time of T ~ 200fs. There is then, a priori, enough time for the ion to fully fill the L shell above the surface. The shortest lifetime for the filling of the K hole in O ions or atoms is that of the neutral atom: $\tau_K \sim 20$ fs; the longest one is that of the Helike ion (τ_K = 300fs for the $^1P_1$ state).It is then likely that the K hole of many of these quasi neutralized ions may be filled above the surface, even if these lifetimes may be lengthened when the ions are re-fed (Zeno effect(2)).

The same kind of spectroscopic features, and of evolution with the ion kinetic energies has been observed, at first order, for O^{7+} on metals and insulators. Moreover with these ions we do not observe, in X ray spectroscopy, any saturation effect for Au, nor for Si or SiO_2, even at the lowest ion kinetic energies(1 or 2 eV/q), but instead we observed a continuous evolution of the spectra. One must however take in mind that the minimum energy gain for O^{7+}(2.6 eV) is comparable to the width of the ion kinetic energy distribution.

The X rays observed at very low energies are made of the characteristic 'outside' and 'touch down' spectra. In the case of Ar^{17+} the 'outside' spectrum is made of the single KL^1 satellite. As soon as the ion touches the surface few (at least 2 or 3) electrons are captured in the M and N shells(1) and these excited states immediately decay through the emission of a fast LMM Auger transition, adding one more electron into the L shell, and leading to a sudden increase of the KL^2 line. The continuous change of the spectra observed above dielectrics with increasing kinetic energies is then only due to a continuous increase of the touch down signal. In the case of O^7 the ion directly fills, at contact its L shell, the so called side feeding (13), quickly increasing the number of L electrons. The evolution of the spectra with decreasing kinetic energies displays an increase of the intensity of the $KL^{1,2,3}$ satellites (mainly emitted by hollow atoms) with respect to that of the $KL^{4,5,6}$ ones. As this $KL^{4,5,6}$ line signs the touch down of the ions as well as the end of the decay of the hollow atoms above the surface, the continuous evolution of the observed spectra displays, in an unknown proportion, the increase of the number of ions touching the surface with increasing kinetic energies, and the evolution of the behavior of the hollow atoms with the travel time (alternate captures and autoionizations).

What happens above the surface and below z_0 mainly depends on the mean charge of the ion during the capture and loss of electrons processes. Below z_0 the ion gains, through the image acceleration, some additional energy before touching the surface. Above dielectrics the ion gains energy through its image acceleration, but also loses energy due to the repellent force induced by the remnant holes on the surface. In the case of Ar^{17+} the ion, as above discussed, is highly charged above metals and dielectrics, and the total 'extra' energy gain(below z_0) is always quite large, as well as above dielectrics the energy lost. The difference between the X ray spectra observed above conductors and insulators is then maximized, as experimentally demonstrated(9). With O^{7+} most of the ions are quasi neutralized and the 'extra' energy gain or loss below z_0 are, by comparison to what happens for Ar^{17+}, minimized, and one cannot really observe, in agreement with the present experimental results, any difference between the evolution of the X ray spectra emitted by the ions above conductors and dielectrics. Similar findings have also been found for Ne^{9+} ions (14). One cannot then deduce any information about a possible backscattering of the O^{7+} ions above dielectrics, as well in X ray as in Auger spectroscopy.

Conclusions

We directly demonstrated that above dielectrics Ar^{18+} ions may be backscattered, and that the capture and loss of electrons proceeds through stepwise waves of capture followed by full autoionization. It is envisaged to go further to study the second step of capture-autoionization, following the filling of the first K hole by looking at the hypersatellite and satellite spectra with Ar^{18+} ions (filling of two K holes),and/ or the K and L X ray cascade. The behavior below z_0 of Ar^{17+} ions(ending the first step of capture–ionization as highly ionized) and O^{7+}(quasi neutralized) has been found to be qualitatively different not allowing any extrapolation of the properties of one of them to the other.

References

(1) J.P.Briand et al. Phys.Rev.Lett. **65**(1990) 159.
(2) J.P.Briand, Comments At.Mol.Phys.**32**(1996) 32.
(3) F.Aumayr et al. Comments At.Mol.Phys.**29** (1994) 275.
(4) F.W.Meyer et al. J.Phys.(Paris) Coll . **50 C1 (**1989) 263.
(5) S.Schippers et al. Phys.Rev.**A 50** (1994)540.
(6) R.Kohrbruck at al. Phys.Rev.**A 45**(1992) 4653.
(7) J.P.Briand et al. Phys.Rev. **A 53** (1996)2925.
(8) J.P.Briand et al. Nucl.Instr.and Meth.**B 154** (1999)166.
(9) J.P.Briand et al. Phys.Rev. **A 55**(1997) R2523.
(10) T.Niedermayr et al. to be published.
(11) J.Burgdorfer et al. Phys.Rev. **A44**(1991)5674
(12) N.Vaek et al. J.Phys.: At.Mol.Opt.Phys. **28** (1995)3523.
(13) L.Folkerts et al.Europhys.Lett.**13**(1990)377.
(14) S.Daveau et al. to be published.

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