ESRF	**Experiment title:** **Double K-shell photoionization of Al and Si from threshold to saturation**	**Experiment number**: HS-2934
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Report:

Introduction

Experimental information concerning the double K-shell ionization induced by impact with electrons [1-2], protons [3], α-particles [4] and heavy ions [5-7] is available for elements $20 \leq Z \leq 50$. Low-resolution and high-resolution measurements were performed. In the first case the technique consists to perform coincidence measurements of the hole transition cascade $1s^{-2} \rightarrow 1s^{-1}2p^{-1}$ - $1s^{-1}2p^{-1} \rightarrow 1s^{0}2p^{-2}$. In the high-resolution approach, the K-shell double ionization cross sections are determined from the relative intensities of the resolved hypersatellite x-ray lines.

In collisions involving charged particles and heavy ions the double 1s ionization is dominated by the direct Coulomb ionization process whereas the double 1s photoionization is related to electron-electron correlation processes. Cross sections for double K-shell vacancy production following photon impact are thus considerably smaller than those corresponding to collisions with heavy ions. As a consequence, intense photon beams such as those delivered by synchrotron radiation sources are needed for the sample irradiation and good peak-to-background and resolution performances should also be evinced by the employed detector system.

Photo-induced K-hypersatellites are thus difficult to observe and experimental data are scarce. By now only results for Ca, Ti and V [8], Ti, Cr, Fe, Ni and Cu [9] and Mo [10] are available. Data reported in [8] and [9] were measured at Spring8 and at the ESRF, respectively, by means of high-resolution, whereas the Mo data [10] were obtained at the APS, using the low-resolution hypersatellite-satellite coincidence technique. Very recently, the double K-shell photoionization of Ne was also investigated at the APS at an excitation energy of 5 keV by recording the KK-KLL Auger-electron spectrum [11].

We report here on high-resolution measurements of the K-hypersatellites of Al and Si. For Al the hypersatellite transition was measured at ten excitation energies ranging from 3186 eV (onset energy) to 5451 eV (saturation regime) whereas for Si data were collected only at two beam energies, namely at 3750 eV (below the threshold) and 5713 eV (saturation).

Experiment

The radiative decay channel of $1s^{-2}$ doubly excited states is governed by the emission of so-called hypersatellite x-ray lines which are shifted towards higher energy relative to the parent diagram lines by about 130 eV for light elements such as Al and Si. TEOP (two-electron-one-photon) transitions [12-14] in which the two K holes are filled simultaneously by two 2p electrons and a single photon is emitted represent a competitive decay channel of two K-vacancy states. However, according to theoretical predictions TEOP transitions are three orders of magnitude less probable than hypersatellite transitions and can thus be neglected.

The measurements were performed at the ESRF beam line ID 21 with a von Hamos Bragg-type curved crystal spectrometer [15]. For the Al measurements, a ADP (101) crystal (2d = 10.642 Å) was employed, whereas for the Si measurements the spectrometer was equipped with a SiO_2 $(1\bar{1}0)$ crystal (2d = 8.5096 Å). The fluorescence x-ray emission of the samples was observed through a 0.2 mm wide rectangular slit positioned in front of the target, in the target-crystal direction. The x-rays diffracted by the crystal were recorded with a 26.8 mm long and 8 mm high position-sensitive back illuminated CCD detector consisted of 1340 columns and 400 rows with a pixel resolution of 20 x 20 μm^2.

The synchrotron radiation beam was monochromatized by means of two 20 Å Ni/B_4C multilayers and residual high-energy photons were suppressed with a Ni mirror. The incident flux on the targets was about $2 \cdot 10^{12}$ photons/s. The Al target consisted of a 1.0 µm-thick metallic foil. For Si, a 1.0 mm-thick single crystal was employed. The energy calibration of the x-ray beam was derived from measurements of the K-absorption edges of S, Ca, Ti and V. The energy calibration of the hypersatellite x-ray spectra was determined from measurements of the Kα diagram transitions of Al and Si for the Al sample and Si and P for the Si one. These measurements were also employed to determine the instrumental broadening of the spectrometer. The latter was found to be well reproduced by Gaussian functions with FWHM of 0.76 eV for Al and 0.55 eV for Si. All x-ray spectra were normalized off-line for the number of incident photons and acquisition time and corrected for the beam intensity profile.

Results

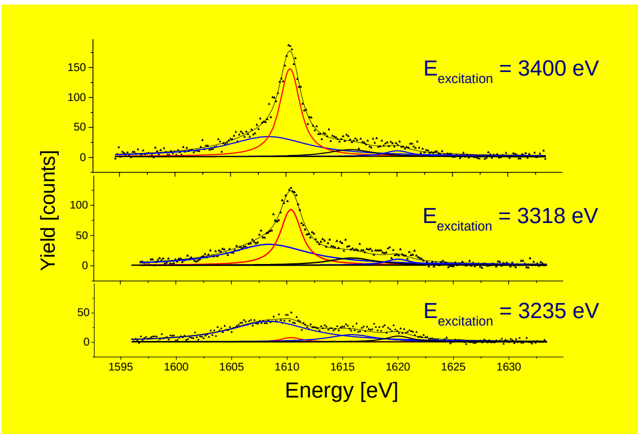

Fig. 1: Evolution of the $K\alpha_2^h$ hypersatellite yield of Al (red line) as a function of the beam energy. The blue curves stand for the $K\beta L^{(2)}$ satellite contamination.

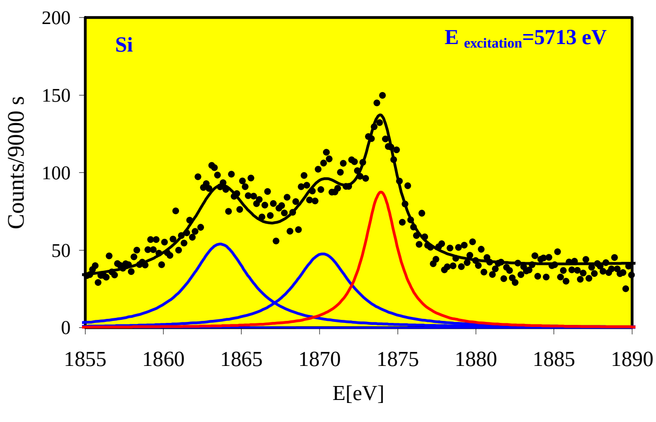

Fig. 2: $K\alpha_2^h$ hypersatellite (red line) and overlapping $K\beta L^{(1)}$ satellites (blue lines) of Si at saturation.

For light elements such as Al and Si, the hypersatellite lines are situated above the K absorption edge. Their already weak intensities are therefore additionally attenuated by the increased self-absorption in the target, which makes their measurements very challenging. For both targets, an additional serious experimental difficulty arose from the overlapping of the KβL satellite lines ($K\beta L^{(2)}$ for Al and $K\beta L^{(1)}$ for Si) with the weak $K\alpha_2^h$ hypersatellites. The problem could be solved by measuring for both targets the L-satellite structures at beam energies below the threshold energies corresponding to the production of the double 1s vacancies. For each beam energy above the threshold, the so-determined L-satellite yields were then corrected for the difference in the 1s photoionization cross section and subtracted from the hypersatellite spectra.

For illustration the $K\alpha_2^h$ hypersatellite lines ($1s_{1/2}^{-2} \rightarrow 1s_{1/2}^{-1}2p_{1/2}^{-1}$ transitions) of Mg and Si observed at three beam energies for Al (Fig. 1) and at 5713 eV for Si (Fig. 2) are shown. Note that the $K\alpha_1^h$ hypersatellite which corresponds to the $1s_{1/2}^{-2} \rightarrow 1s_{1/2}^{-1}2p_{3/2}^{-1}$ spin-flip transition is forbidden by the E1 selection rules in the LS coupling scheme and cannot therefore be observed for light elements such as Mg and Si for which the LS coupling scheme prevails. As shown in Fig. 3, for Al the evolution of the hypersatellite intensity as a function of the beam energy could be well fitted, using the Thomas model [16].

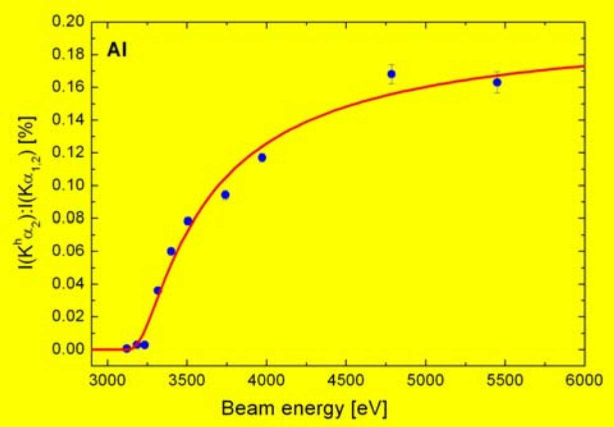

Fig. 3: Evolution of the $K\alpha_2^h$ yield as a function of the beam energy. The red curve represents the fit to the experimental data of the Thomas function describing the transition from the adiabatic to the saturation regime.

Very precise results could be obtained for the energies and widths of the hypersatellite transitions. The energies (1610.38 ± 0.05 eV for Al, 1873.92 ± 0.06 eV for Si) were found to be in satisfactory agreement with the values obtained from MCDF calculations, whereas for the linewidths, significantly bigger values (1.70 ± 0.16 eV for Al and 2.42 ± 0.20 eV for Si) than those predicted by theory (1.15 eV and 1.34 eV) were obtained. However, the most interesting result concerns the probabilities for the production by photoionization of double 1s vacancies which are for both Al ($1.92 \cdot 10^{-3}$) and Si ($1.43 \cdot 10^{-3}$) about one order of magnitude bigger than the values expected from calculations based on the shake model ($1.7 \cdot 10^{-4}$ and $1.4 \cdot 10^{-4}$) [17]. A similar big discrepancy was found in our previous hypersatellite project concerning Mg. This discrepancy is probably due to the TS1 process (knock-out of the second 1s electron by the outgoing K photoelectron) which is not considered in the shake calculations based on the sudden approximation model. It can be noted that similar discrepancies have been observed by other groups for heavier elements [8-11].

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