ESRF	**Experiment title:** Double K-shell photoionization of Mg, Al and Si from threshold to saturation	**Experiment number**: HS-2404
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Report:

Introduction

Hollow atoms are atoms in which the most inner-shell is empty, while the outer shells remain intact. The easiest way to produce double 1s vacancies consists to bombard the neutral atoms with charged particles. Experimental information concerning the double K-shell ionization induced by impact with electrons, protons, α-particles and heavy ions is available for elements $20 \leq Z \leq 50$. Production of double K-vacancies by photon impact is more difficult to achieve because atomic inner-shell photoionization is a rather soft process, in a perturbation sense, in which a single photon can be assumed to interact with a single bound electron. As a consequence, double K-shell photoionization can only result from electron-electron correlation effects such as shakeup and shakeoff processes.

K-shell shake probabilities following 1s photoionization are, however, very small. Photoinduced K-hypersatellite transitions are thus difficult to observe and experimental data are scarce, except for He which has been extensively studied [1]. Very few heavier elements have been investigated. By now only results for Ca, Ti and V [2], Ti, Cr, Fe, Ni and Cu [3] and Mo [4] are available. Except the results reported in [5] that were obtained with an x-ray tube, all measurements were performed, using intense synchrotron radiation for the irradiation of the samples. Data reported in [2] and [3] were measured at the Japanese synchrotron source Spring8 and at the European Synchrotron Radiation Facility (ESRF), respectively, by means of high-resolution, whereas the Mo data were obtained at the Advanced Photon Source (APS, USA), using the low-resolution hypersatellite-satellite coincidence technique. Very recently, the double K-shell photoionization of Ne was also investigated at APS at an excitation energy of 5 keV [6]. However, since for light elements the hypersatellite x-rays whose energies lie above the K absorption edges are strongly attenuated by the self-absorption in the target and are thus extremely weak, these Ne measurements were performed by means of Auger-electron spectroscopy.

We report on high-resolution measurements of the K-hypersatellite fluorescence x-ray emission of Mg, Al and Si samples. To our knowledge our experiment represents the first attempt to measure photoinduced K-hypersatellites of low-Z targets. Good data were obtained for Mg for which the hypersatellite transition of interest could be measured at different excitation energies ranging from threshold to saturation whereas for

Al and Si, due to a lack of beamtime and some severe problems encountered with the CCD detector of the crystal spectrometer, data could be collected at only one excitation energy and with poor statistics. In addition at the higher beam energies used for the irradiation of the Al and Si samples, the overall background was more important.

Experiment

The measurements were performed at the ESRF beam line ID 21 by means of high-resolution x-ray spectroscopy, using a von Hamos Bragg-type curved crystal spectrometer [7]. The latter was installed downstream of the STXM chamber to which it was connected through a 200 cm long evacuated pipe. For the Mg measurements, a TlAP(001) crystal (2d = 25.772 Å) was employed, using the 2nd order of reflection, whereas for the Al and Si measurements the spectrometer was equipped with a ADP(101) crystal (2d = 10.642 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 10^{12} photons/s. The Mg and Al targets consisted of metallic foils, with thicknesses of 3.3 μm and 1.0 μm, respectively. 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, Cl, Ca, Ti and Cr. The energy calibration of the von Hamos spectrometer was performed by measuring the $K\alpha_{1,2}$ diagram transitions of the three samples. These measurements were also employed to determine the instrumental broadening of the spectrometer. All x-ray spectra were normalized off-line for the number of incident photons and acquisition time and corrected for the beam intensity profile. In the von Hamos slit-geometry different regions of the crystal surface see different parts of the target so that the shape of the measured spectrum is slightly affected by the spatial distribution of the beam intensity on the target.

For each photon beam energy, the following sequence of measurements was performed:

- determination of the beam intensity profile on the target
- short measurement of the Kα x-ray spectrum ($1s^{-1} \rightarrow 2p^{-1}$ transition) (typically 400 images with an exposure time of 1 s. per image)
- long measurement of the Kα hypersatellite x-ray spectrum ($1s^{-2} \rightarrow 1s^{-1}2p^{-1}$ transition) (typically 1000 images with an exposure time of 10 s. per image)
- repetition of the short Kα measurement to probe the stability of the beam and reproducibility of the experimental set-up.

Results

For Mg, the diagram and hypersatellite x-ray spectra were measured at 9 different photon beam energies (2746 eV, 2761 eV, 2796 eV, 2846 eV, 2896 eV, 2996 eV, 3096 eV, 3522 eV and 3947 eV). At 2746 eV, no significant hypersatellite yield was observed, whereas at 2761 eV a yield ratio of $1.1(2)\cdot10^{-4}$ between the hypersatellite and diagram lines was found. The onset energy for the production of double 1s vacancy states in Mg lies therefore between these two energies, i.e. at about 2750 eV. Above the threshold energy, a fast rise of the hypersatellite intensity with the excitation energy is observed, becoming less and less pronounced for increasing beam energy so that the hypersatellite yield tends toward a saturation value, as predicted by the theory. At the highest incident energy (3947 eV) the measured hypersatellite-to-diagram line yield ratio was found to be $1.92(12)\cdot10^{-3}$, i.e. a value about one order of magnitude bigger than theoretical predictions from shake calculations based on the sudden approximation model.

From the fits, the average energy shift of the hypersatellite with respect to the parent diagram line was found to be 114.28(3) eV, in good agreement with the value of 114(1) eV obtained in previous measurements performed with fast heavy ions [8]. As the width of a transition is given by the sum of the natural widths of the initial and final states, the width $\Gamma_{K\alpha}{}^{h}$ of the hypersatellite transition should be equal to $3\cdot\Gamma_K + \Gamma_L$ if one assumes that the width Γ_{KK} of a double 1s vacancy state is equal to two times the width Γ_K of a single vacancy state. Using this approximation and the widths Γ_K (0.33 eV) and Γ_L (0.03eV) reported for Mg by Campbell and Papp [9], a value of 1.02 eV is expected that is significantly smaller than the value of 1.92(10) eV

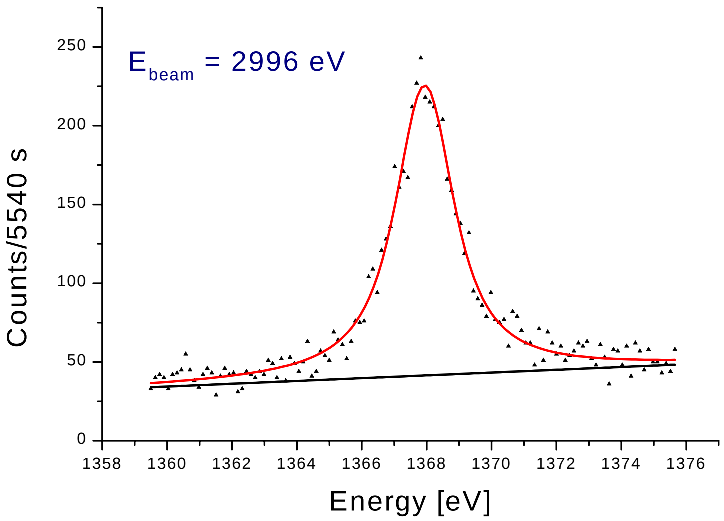

Fig. 1: Kα_2^h hypersatellite transition of Mg observed at a beam energy lying about 500 eV above the threshold energy for the double 1s photoionization.

deduced from the fits of our data. This intriguing discrepancy is too big to be explained by the broadening of the transition resulting from the shake-induced additional M-shell ionization.

For illustration the Kα_2^h hypersatellite line of Mg ($1s_{1/2}^{-2} \rightarrow 1s_{1/2}^{-1}2p_{1/2}^{-1}$ transition) observed at a beam energy of 2996 eV is shown above. Note that the Kα_1^h hypersatellite ($1s_{1/2}^{-2} \rightarrow 1s_{1/2}^{-1}2p_{3/2}^{-1}$ transition) which corresponds to a 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 for which the LS coupling scheme prevails.

Due to a smaller intensity than expected of the Mg hypersatellites and a faster increase above the threshold of the hypersatellite yields requiring more beam energy steps than anticipated, only 3 shifts were left for the Al and Si measurements. The latter were therefore performed at a single beam energy, namely at 4794 eV for Al and 5769 eV for Si. During the replacement of the TlAP(001) crystal by the ADP(101), the spectrometer chamber was probably opened too early so that some water vapour or other unknown impurity condensed on the still cold CCD surface. As a result a large part of the CCD area was completely inefficient, the low-energy photons being absorbed by the liquid deposed on the surface. In addition to the resulting strong reduction of the detector efficiency, the spectra were affected by an inhomogeneous background that made the hypersatellite fits poorly reliable. In our opinion, these data are hardly usable and the measurements should be repeated.

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