| ESRF | **Experiment title:** Two-electron one-photon transitions in Al and Si following double K-shell photoionization. | **Experiment number**: HE-3194 |
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| **Beamline**: | **Date of experiment**: from: 22.10.2009 to: 27.10.2009 | **Date of report**: 24.06.2011 |
| **Shifts:** 12 | **Local contact(s)**: Dr. Jakub Szlachetko | *Received at ESRF:* |

**Names and affiliations of applicants** (* indicates experimentalists):

J. Hoszowska*, Physics Department, University of Fribourg, 1700 Fribourg, Switzerland
J.-Cl. Dousse*, Physics Department, University of Fribourg, 1700 Fribourg, Switzerland
Y. Kayser*, Physics Department, University of Fribourg, 1700 Fribourg, Switzerland
W. Cao*, Physics Department, University of Fribourg, 1700 Fribourg, Switzerland
S. Nowak* Physics Department, University of Fribourg, 1700 Fribourg, Switzerland
J. Szlachetko*, European Synchrotron Radiation Facility, F-38043 Grenoble, France
M. Kavcic*, J. Stefan Institute, SI-1000, Ljubljana, Slovenia
P. Jagodzinski*, Institute of Physics, Jan Kochanowski University,25-400 Kielce, Poland

## Report:

## Indroduction

The aim of the study was to observe for the first time the correlated two-electron one-photon (TEOP) transitions following single-photon K-shell double ionization and to determine the $K\alpha^h$ to $K\alpha\alpha^h$ branching ratios and the TEOP transition energies.

The K-shell double photoionization (DPI) is one of the most sensitive probes of inner-shell electron correlations [1,2]. The radiative decay of the K-shell double vacancy states proceeds mainly via the one-electron-one-photon (OEOP) process which corresponds to the $K\alpha^h$ hypersatellite transition ($1s^{-2} \rightarrow 1s^{-1}$-$2p^{-1}$). In an alternative decay channel, the two-electron one-photon (TEOP) transition $K\alpha\alpha^h(1s^{-2}\rightarrow 2s^{-1}2p^{-1})$, the two K-shell vacancies are filled *simultaneously* via a correlated two-electron jump. In comparison to the Kα diagram fluorescence lines the TEOP transition rates are smaller by a factor of ~$10^6$-$10^7$. As a consequence, measurements of TEOP transitions are very challenging. First experimental evidence for TEOP was found by Wölfli et al. [3]. However, the multiple-ionization in heavy-ion collision experiments makes comparison with theory often inconclusive. On the theoretical side, predictions for the TEOP transition rates differ up to a factor of four [4-9]. In this context, the photon impact data provide a more stringent test for calculations.

### Experiment

The TEOP measurements for solid Al and Si were carried out using the recently developed wavelength dispersive x-ray spectrometer (WDS) of the ID21 beamline [10]. The high-efficiency and good energy resolution of the spectrometer were prerequisite for the experiment. The beam was produced by two undulators and monochromatized using the double Ni/$B_4C$ multilayer monochromator. The upper harmonics were rejected with Ni-coated mirrors. The beam was focused down to ~10x10μm$^2$ by means of the KB device, and the micro-focused incident photon flux was ~2x$10^{12}$ photons/s. The photon energy in the region of the K-shell double photoionization cross-section maxima of 4.620 keV was chosen.

The WDS is based on a polycapillary optics for x-ray fluorescence collection, a flat crystal and a flow gas x-ray detector. The latter are mounted on a theta-2theta rotation stage, where theta stands for the crystal Bragg angle of diffraction. The spectrometer was equipped with a Si(111) crystal for the Al and a Ge(220) crystal for the Si measurements, respectively. A self-supported metallic foil of Al (9.4 µm-thick) and a 1 mm-thick crystal of Si, were employed. The Al and Si sample purity was 99.999 %. The samples were tilted so that both the incident beam direction and the direction of observation were at an angle of 30 degrees to the sample surface normal. The energy calibration of the WDS was based on the recommended energies and assigned to the measured Cl, Pd, Ag, Sn, K, and Sc diagram transitions. These also served to determine the full width at half maximum (FWHM) of the Gaussian instrumental response function which was found to be in the 7-10 eV range. The x-ray spectra were collected in successive theta-2theta scans of ~0.5-1 h each, with total acquisition times of ~51 h for Al and ~17 h for Si. For normalization purposes the photon flux was recorded at the beginning and the end of each scan, and the stability of the experimental setup was monitored with the Pd $L\beta_{3,4}(L_1\text{-}M_{3,2})$ and K Kβ emission lines for the Al and Si TEOP x-ray spectra measurements, respectively.

**Results**

To the best of our knowledge, this experiment represents the first observation of the TEOP transitions resulting from single photon impact. For illustration, the x-ray spectrum for Si is depicted in Fig 1.

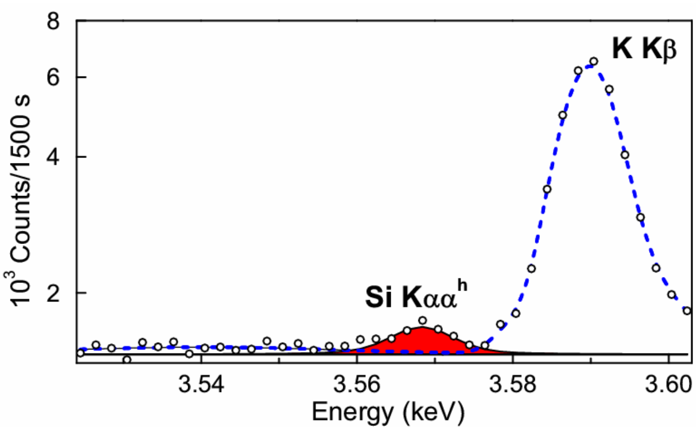


Fig. 1. *Two-electron one-photon transition of Si (filled area) and the K Kβ x-ray spectrum due to trace impurities (dashed line).*

The TEOP transitions of Al and Si are shown in Fig.2. Since for light elements the L-S coupling scheme prevails the $^3P_1 \rightarrow {}^1S_0$ transition is dipole-forbidden, and thus only the $K\alpha_2\alpha_3^h$ ($^1P_1 \rightarrow {}^1S_0$) transition was observed.

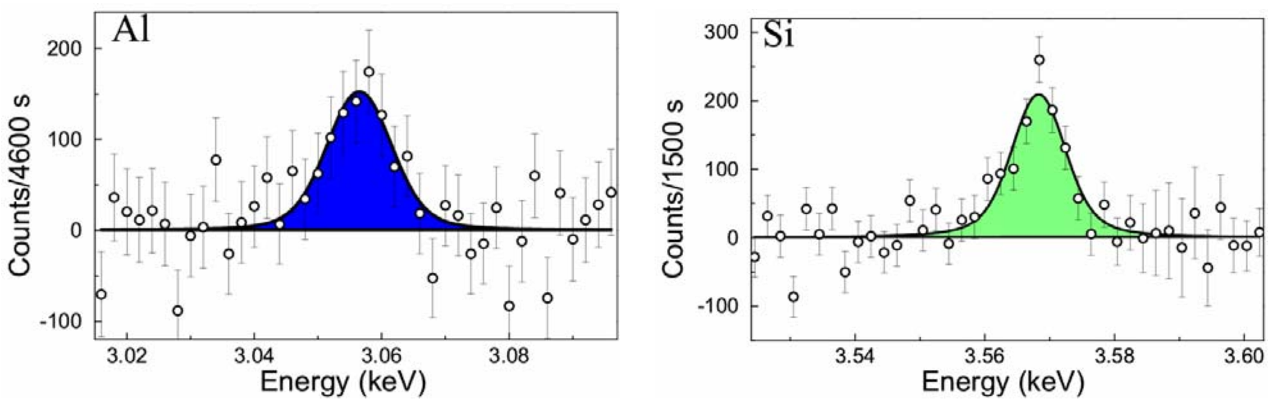


Fig. 2. *Two-electron one-photon transitions of Al and Si.*

As it was not possible to measure both the TEOP and the $K\alpha^h$ hypersatellite transitions with the same crystal, the branching ratios were derived from the measured intensities of the close lying diagram lines of K Kα and Cl Kα for Al and the Sc Kα and K Kβ for Si, the K-shell single photoabsorption cross sections and the known DPI cross sections [2]. The precision of the method, assesed from several K and L x-ray emission line yields measurements, was found to be better than 5%.

The $K\alpha^h$ to $K\alpha\alpha^h$ branching ratios of 2115(403) for Al and 2610(370) for Si, and the TEOP transition energies of 3056.5(9) eV for Al and 3568.3(4) eV for Si were determined and compared to available theoretical predictions and data from heavy-ion collision experiments. The TEOP transition energies were found to be in good agreement with the calculated MCDF values [11]. The average-field approximations and MCDF predictions, however, do not adequately describe the electron-electron interaction in the initial and final states of the TEOP transitions and underestimate the branching ratios. The present branching ratios compare best to perturbation theory predictions [9] and configuration interaction calculations [8].

First results were presented at the 37th International Conference on Vacuum UltraViolet and X-Ray Physics, July 11-16, 2010, Vancouver, Canada and a manuscript was submitted recently to Phys. Rev. Lett.

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