	Experiment title: **Near-threshold argon KL satellite structures studied by high resolution x-ray spectroscopy**	**Experiment number**: MI-455
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Report:

Introduction

Due to correlation effects between core-electrons, the 1s photoionization of atoms may be accompanied by the ejection into the continuum of a second inner-shell electron (shakeoff) or by the promotion of the latter in an unfilled upper level (shakeup). An efficient method to investigate these multielectron effects consists to observe with high-resolution the K x-ray fluorescence emission spectrum of the target. As a result of the double core-vacancy configuration characterizing the initial states, such spectra evince indeed satellite structures whose shape and intensity vary as a function of the excitation energy. The aim of our experiment was to perform a detailed investigation of the Ar K+L double excitation in the threshold energy region by observing the evolution of the shape and intensity of the $K\alpha L^1$ x-ray satellite structures corresponding to the $1s^{-1}2s^{-1}\rightarrow 2p^{-1}2s^{-1}$ and $1s^{-1}2p^{-1}\rightarrow 2p^{-2}$ radiative transitions.

Experiment

The $K\alpha$ x-ray emission spectrum of Ar was measured with our high-resolution von Hamos bent crystal spectrometer [1] equipped for the present experiment with a 5 x 10 cm^2 $(1\bar{1}0)$ quartz crystal plate bent to a radius of 25.4 cm. The spectrometer was installed at the beamline ID-21 downstream from the microscope chamber to which it was connected through a ~180 cm long evacuated pipe and a 25 μm thick Kapton window. The target consisted of a 4 mm high cylindrical cell filled with 0.9 bar argon gas. In order to get the highest possible beam intensity, a 2 mm pinhole was employed and for each beam energy the monochromator was adjusted to correspond to the top of the rocking curve. The extended beam spot did not affect the resolution of the spectrometer because the target was viewed by the crystal through a narrow tantalum slit (width of 0.2 mm) located in front of the gas cell. The instrumental broadening of the spectrometer was determined from the observed linewidth of the Ar $K\alpha L^0$ diagram line excited by impact with 3206 eV photons for which only single K-shell ionization is energetically possible and consequently no broadening by unresolved M-satellites occurs. For each beam energy, a short measurement of the whole Ar Kα spectrum was performed consisting of typically 500 images with an exposure time of 1 s per image. Such a spectrum is shown for illustration in Fig. 1. Then the $K\alpha L^1$ satellite region of interest (the weak structure appearing around 2977 eV in Fig. 1) was measured in longer scans (exposure time of 10 s per image). In this case, 1000-3000 images were collected, depending on the beam intensity ($2\cdot10^{11}$-$5\cdot10^{11}$ ph $s^{-1}mm^{-2}$) and the double excitation cross section. Beam energies of 3205, 3206 and 3207 eV corresponding to the 1s single ionization threshold region, 3455 eV and 3457 eV (onset for simultaneous 1s2p double excitation), 3500, 3510 and 3512 eV (onset for sequential 1s2p double excitation) as well as energies of 3525, 3550, 3575 (threshold for simultaneous 1s2s double ionization), 3600, 3700, 3800 and 4000 eV were employed. The energy calibration of the monochromator was adjusted by measuring the K absorption edge of argon (3205.9 eV) and by observing with the von Hamos spectrometer the line corresponding to the elastic scattering in the target of the 3206 eV beam. In order to compare our results with theoretical predictions based on the sudden approximation model, an additional measurement was performed at a higher energy (6540 eV). For this last part of the experiment, a new calibration of the beam energy was performed, using the K-edge of Mn at 6539.0 eV. The $K\alpha L^1$ satellite structures observed at excitation energies of 3510, 3550 and 3700 eV, respectively, are shown in Fig. 2. Although the depicted features correspond to less than 30% of the data collected at these energies, one sees that the yields of the L satellites depend strongly on the excitation energy and, to a smaller extent, that the profile and centroid of the bump also vary with the beam energy.

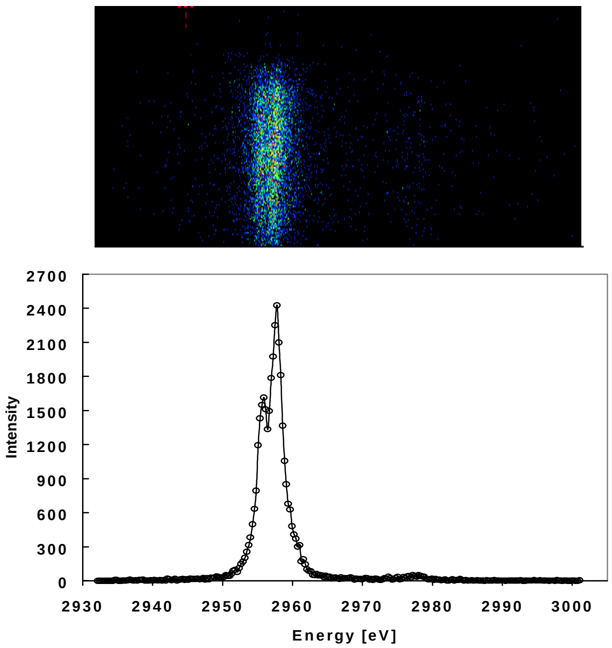

Fig. 1: two-dimensional intensity pattern corresponding to the Kα x-ray spectrum of argon observed with the von Hamos spectrometer. The projection of the image on the dispersion axis results in the energy spectrum shown in the lower part of the figure. The small bump above 2975 eV corresponds to the $K\alpha L^1$ satellite structure of interest.

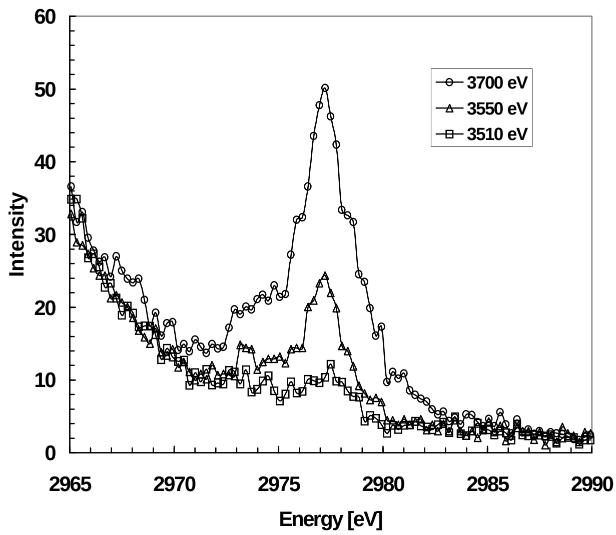

Fig. 2: Ar $K\alpha L^1$ satellite structures observed at three different beam energies. As expected, the satellite yields were found to increase with growing excitation energies. The complex shape of the structures is due to the fact that the satellite transition consists of many overlapping components.

Data analysis and results

The analysis of the observed K x-ray spectra is in progress. The analysis is performed by means of a least-squares-fitting computer program employing Voigt profiles to fit both the $K\alpha L^0$ diagram lines and $K\alpha L^1$ satellites. The convolution of the Gaussian instrumental response of the spectrometer with the Lorentzian function corresponding to the natural line shape of x-ray transitions results indeed in so-called Voigt profiles. As the observed satellite structures which consist of many overlapping components are broad and asymmetric, they are fitted with several juxtaposed Voigt profiles whose intensities, widths and positions are based on extensive multiconfiguration Dirac-Fock (MCDF) calculations [2].

The fitted satellite yields reflect the distribution of the L spectator holes at the moment of the K x-ray emission and not the initial distribution following the 1s photoionization which is of interest for the determination of the shake probabilities [3]. Processes such as radiative, Auger, Coster-Kronig or super Coster-Kronig transitions occuring prior to the K x-ray emission can indeed modify the number of holes created by the shake process. For KL satellites the electron rearrangement is governed by LMX Auger transitions and L radiative transitions. In order to extract the needed primary vacancy distributions from the observed satellite yields, statistical scaling procedures which account for these rearrangement processes have thus to be applied.

From the corrected satellite yields, the probabilities for producing via shakeup and shakeoff processes K+L double vacancy states will then be determined. The results will be compared to theoretical predictions based on the model of Thomas (adiabatic excitation limit) and the shake model (sudden approximation limit). Furthermore, a better understanding of the dynamics of the processes contributing to the double K+L excitation will hopefully be gained from the observed evolution of the shape of the satellite spectrum as a function of the excitation energy.

References

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