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Photon-photon delayed-coincidence spectroscopy as a tool probing the cascade transitions produced after the Ar 2*p* excitation

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Keywords: synchrotron radiation, photon-photon delayed-coincidence technique, argon

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Many investigations have been devoted to obtain information on the electronic excitation and relaxation of the atoms and molecules. In particular the 2*p* excitation and relaxation of atomic Ar in the energy region between 240 and 255 eV have been studied by several different techniques, including photoabsorption [1], photoionization [2], electron-energy-loss spectroscopy [3], threshold electron–Auger electron coincidence technique [4], and UV-VIS fluorescence spectroscopy [5]. Photoabsorption and electron-energy loss spectra of Ar show pronounced structures below the Ar 2*p*-ionization thresholds that are attributed to resonant excitations of a *2p* electrons into low-lying *ns* and *nd* Rydberg states [1,3]. These core-excited states decay predominantly via resonant Auger transitions that serve as a start for relaxation processes occurring through the emission of fluorescence light [5]. But, the emission attributed to ionic states may not be directly formed by primary excitation processes. Therefore, the occurrence of cascade transitions has been proposed to play an important role in electronic relaxation [5]. This, however, complicates the interpretation of the fluorescence spectra. Fortunately, the coincidence measurements allow to avoid these problems and to provide more insight into the dynamics of the corresponding processes.

In the present study, observation of coincidences between fluorescence photons from the decay of the 2*p* inner shell excited argon in the 240-255 eV energy range was performed. The experiments were carried out at the low-energy branch of the Gas Phase Photoemission beamline at the Elettra synchrotron radiation facility utilizing the photon-induced fluorescence spectroscopy (PIFS) [6]. The branch line operated in the photon energy range 14-280 eV. For purposes of these studies a dedicated fluorescence detection system was constructed at Gdańsk University of Technology and transferred to Elettra Gas-phase beamline. It consisted of a beamsplitter and two optical channels, each incorporating interference filter followed by a photomultiplier tube (Figure 1). Outside the vacuum chamber, this system allowed to divide fluorescence signal into two parts with the aid of a beam splitter, which was selected according to the wavelength range of the studied emission. The reflected part of the split beam went through a color filter before being detected by a photomultiplier tube (PMT). A filter with the band pass was most often used in this arm of the experiment. The transmitted part of the light beam was filtered with a narrow-band interference filter before it was detected by a PMT tube. The signals from the two PMTs were fed into a time-to-digital converter system and coincidences between the two photon signals were searched after the completion of the measurement by using the data analysis program Igor.

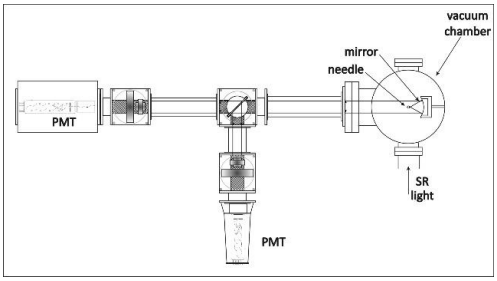

Figure 1. Scheme of detection system.

Here, we have measured fluorescence yields of the 4d′ ^{2}F→4p′ ^{2}F (335 nm) and 5s′ ^{2}D→4p′ ^{2}F (393 nm) decay (see Figure 2) lines and of wideband 380-540 nm emission arising at the 2*p* inner shell excitation of argon. To identify the spectral features we compared these yields with the total ion yield. In consequence, a state-selective behavior was found for those fluorescence excitation spectra. Thus, the coincidence measurements have been performed for the 335 and 459 nm and 393 and 459 nm photons combinations (Figure 2) at various photon energies corresponding to different resonances in argon. For those cascade transitions the coincidence charts and life times of the upper states have been obtained.

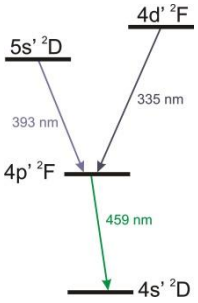

Figure 2. Schematic diagram of cascade transitions measured after the Ar 2*p* excitation.

Acknowledgments: This work was in part financially supported under the CALIPSO contract.

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