Two-Photon Two-Colour Ionization of Rare Gases

M. Meyer, D. Cubaynes, S. Düsterer[1], P. Radcliffe[1], A. Azima[1], H. Redlin[1], J. Feldhaus[1], J. Dardis[2], K. Kavanagh[2], H. Luna[2], J. Pedregosa Gutierrez[2], P. Yeates[2], E.T. Kennedy[2], J.T. Costello[2], A. Delserieys[3], C.L.S. Lewis[3], R. Taïeb[4] and A. Maquet[4]

LIXAM, Centre Universitaire Paris-Sud, Bâtiment 350, F-91405 Orsay Cedex,
[1]HASYLAB at DESY, Notkestrasse 85, D-22607 Hamburg, Germany
[2]NCPST, Dublin City University, Dublin 9, Ireland
[3]IRCEP, Queen's University Belfast, BT7 1NN, United Kingdom
[4]LCP-MR, Université Pierre et Marie, 11 Rue Pierre et Marie Curie, F-75231 Paris Cedex 05, France

In the current phase of our studies of photoionization processes in rare gases, induced by the simultaneous interaction of two photon beams with the atomic target, we have successfully combined the VUV beam delivered by FLASH with pulses from the synchronized optical femtosecond laser. For these experiments, the FEL was operated at 25.5 and 13.8 nm with a repetition rate of 5 Hz in a single bunch mode with typical pulse energies of 10 - 20 µJ and pulse durations of about 20 fs. The optical laser delivered light pulses (800 nm, 20µJ, 120 fs), which were introduced in a collinear geometry into the experimental chamber. Both beams cross an effusive gas jet in the acceptance volume of a magnetic bottle electron spectrometer (figure 1). The high intensity of the FEL, combined with the high collection efficiency of the spectrometer (close to the full solid angle), enables us to record electron spectra for each individual pulse [1]. In the electron spectrum, additional structures appear on both sides of the main photolines, when both The VUV and optical pulses overlap in space and time. These "sidebands" are caused by the common interaction of the VUV and optical pulses and can be described by the above threshold ionization (ATI) process. Their analysis permits us to obtain a very precise characterization of the experimental conditions, in particular the temporal stability of the FEL with respect to the optical laser, while also providing direct access to the electron interaction and correlations in the continuum in the presence of a strong electromagnetic field. Following the first experiments, where we used a picosecond laser in order to minimize effects of the temporal fluctuations of the FEL with respect to the optical laser [2], we have now extended our studies of two-colour ATI to the femtosecond time regime.

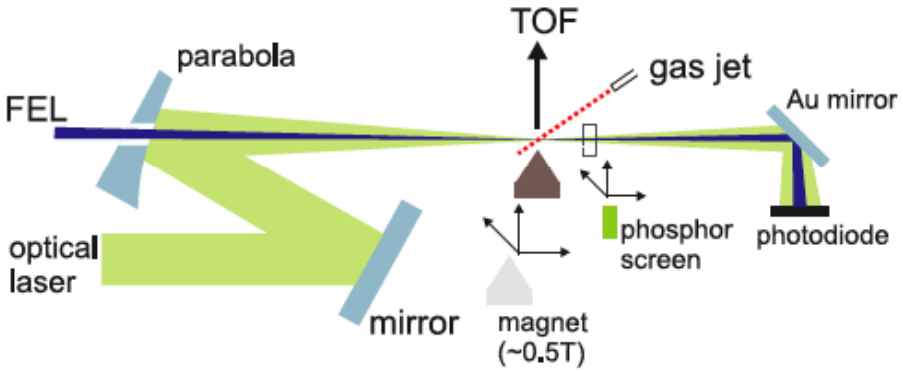

Figure 1: Schematic drawing of the experimental set-up used for the two-colour photoionization studies on rare gases. The analysis of the kinetic energy of the emitted photoelectrons is performed by measuring the time-of-flight (TOF) of the electrons in a magnetic bottle type spectrometer.

The femtosecond optical laser provides a much higher field in the interaction region than the field obtained with the picosecond laser (about 2×10^{12} W/cm^2, when the optical laser is focussed down to a beam diameter of about 50 µm). As a result, much stronger sidebands are observed and, for the

experiments on atomic Xe, it was even possible to observe *the absorption of several optical photons* during the ionization process (figure 2, left-hand side). Up to four sidebands, i.e. absorption of up to four optical photons, are seen in the spectrum for perfect overlap of both pulses. Due to the monochromaticity of the FLASH beam, the higher sidebands are particularly interesting for the comparison with theory, since they permit the study of the interaction of the atom with two or more photons in a direct way. The performed theoretical simulations are based on the solution of the Time-Dependent Schrödinger Equation (TDSE) [3] and are in excellent agreement with the experimental features (figure 2, right-hand side).

In addition, the observation of the sidebands allow for a quantitative analysis of the temporal resolution of the complete pump-probe set-up. The cross correlation curve yields a total width of about 600 fs, which corresponds mainly to the temporal jitter of the FEL pulses with respect to the optical laser. Therefore this value indicates therefore the effective temporal resolution of the set-up without any further determination of the relative delay between the individual pulses. This is the value that would be directly introduced to all pump-probe experiments at FLASH where the final signal is obtained by averaging over many FEL pulses. In order to do better than 600 fs, single shot spectra can be recorded, time-stamped and post-analyzed. The intensity of the sidebands and/or the number of the observed sidebands are a direct indication of the temporal position of the FEL pulse underneath the optical pulse. Thus, the sideband signal recorded shot-by-shot in combination with another pump-probe experiment, can be used to bin the experimental data according to the relative temporal delay between the FEL and the optical pulses. After comparison with the theoretical simulation for different intensities of the optical and VUV fields, a temporal resolution of less than 50 fs was deduced from the present experiment for cases when both pulses overlap [4].

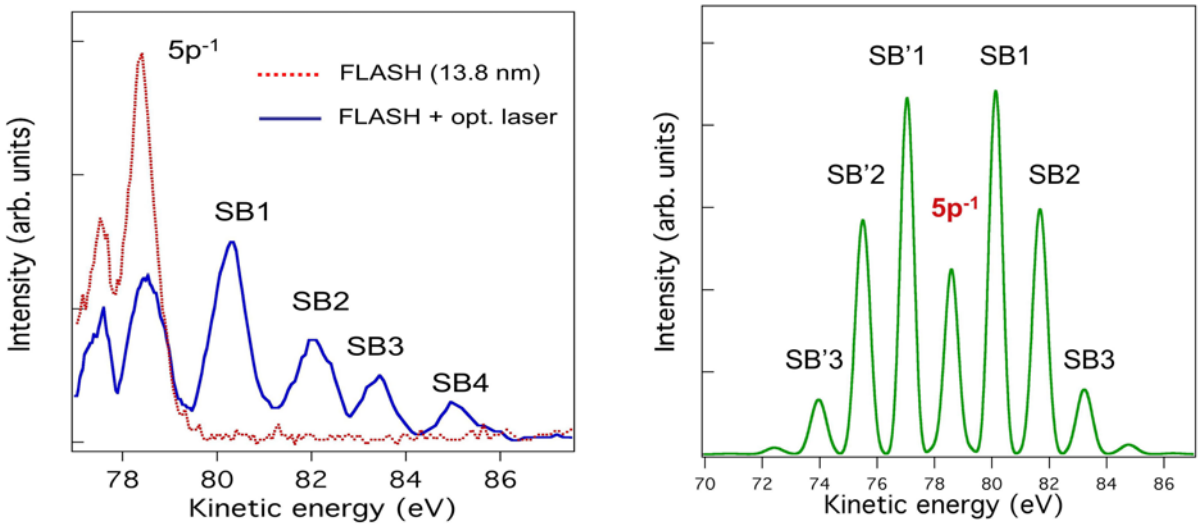

Figure 2: (left-hand side) Single-shot photoelectron spectrum of atomic Xe recorded for perfect overlap between the pulses of FLASH and the optical laser (blue line) and for FLASH pulses only (red dotted line). Only the high energy side of the Xe 5p photolines is shown. (right-hand side) Simulation of the experimental spectrum obtained by solving the Time Dependent Schrödinger Equation (TDSE) for $6x10^{11}$ W/cm^{-2}.

References

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