Photoionization Experiments with the Ultrafast EUV Laser 'FLASH' – Free Electron Laser in Hamburg

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Abstract. The advent of mirror-less Free Electron Lasers emitting polarised and coherent 'laser-like' beams of high peak (> 1GW) and average (up to 100mW) powers in the extreme-UV (EUV) and X-ray bands of the electromagnetic spectrum heralds a new era in the study of the photoelectric effect. The unprecedented photon flux (~10^{13} photons per pulse) opens up to scrutiny processes with cross sections considered hitherto unfeasibly small to probe with conventional EUV sources such as synchrotrons and laser plasmas. The peak intensity of the focussed pulse train (>10^{13}W/cm^2), combined with the high photon energy, ports non-linear optics and spectroscopy into a regime where inner shell electrons can become the predominant mediator of the photon matter interaction. Few photon, few electron photoionization processes are made amenable to study for the first time and the wavelength tunability of the FEL permits resonances to come into play. In combination with ultrafast optical lasers, pump-probe experiments on atoms and molecules where both fields are of comparable high intensity but orders of magnitude different in photon energy become possible. In mid 2005 the 2nd phase of the Free Electron Laser project at DESY, Hamburg (FLASH) opened to users. In what follows I will attempt to illustrate at least some of the impressive progress that has been made by very brief descriptions of just a few of the pathfinder experiments that the growing Atomic and Molecular physics community at FLASH has undertaken in the intervening two years.

1. Introduction

The field of extreme-UV (EUV) science and technology has seen a rapid growth in development and a concomitant widening of interest over the last few decades, motivated by both a fundamental interest in EUV-matter interactions but also by the prospects the field offers for to address bottlenecks in impending nano-manufacturing (e.g., EUV Lithography) and bio-imaging (e.g., *in vitro* targets). The key effect of the interaction of EUV radiation with matter is *photoionization* and it has been a treasure trove (at times a Pandora's box) of intriguing and occasionally bemusing, but always beautiful, many electron interactions and quantum interferences. Experiment and theory have worked side by side in a symbiotic drive to deliver complete experiments along with a deep understanding of the role of many electron effects in photoionization [1]. In all of these studies, the EUV field has been weak and so each atom or molecule absorbs one, and only one, photon.

The competing field of ionization of atoms and molecules in intense infrared and visible laser fields has revealed many new dynamical processes such as multi-photon, above-threshold and optical-field ionization [2] as well as short wavelength coherent radiation via high harmonic generation or HHG [3]. Of course, the interesting question arises, *what happens if the driving field is itself of very*

high optical frequency, say >10^{16} Hz ? Although HHG can provide coherent radiation of moderate average and peak intensity at high frequency, even highly optimised systems are really only on the threshold of driving non-linear physics at short wavelengths [4]. Plasma based lasers have also tried to meet this grand challenge, starting with the first observation of population inversion/ gain in a recombining plasma in the 1970s [5], followed by the first convincing demonstration of a practical collisionally-excited EUV laser at Livermore [6] in the mid 1980s. Despite heroic efforts to improve these systems over the intervening decades by e.g., injection seeding [7], they have not resulted (as yet) in high average power, GW level pulses.

In this regard the opening up of phase two of the EUV Free Electron Laser at DESY, Hamburg, now known as 'FLASH' heralds a new era in the study of intense EUV laser-matter interactions and indeed for the photoeffect itself. The EUV output of FLASH possesses unprecedented intensity and coherence, bringing non-linear optics and spectroscopy into a regime where inner shell electrons can mediate the fundamental photon-matter interaction. For the first time, atomistic systems, bathed in intense and coherent ultrahigh frequency electromagnetic fields, can be uniquely studied using photon, photoion and photoelectron spectroscopies. Experiments on dilute samples and/or species with ultralow photoionization cross sections, few photon processes in few electron systems, two colour (EUV + IR/EUV + EUV) pump-probe experiments on a femtosecond (even attosecond) time scale are all made possible with the advent of ultrafast EUV and soon the XFEL [8] and LCLS [9] X-ray FELs.

Starting operation at a fundamental wavelength of 32 nm in mid 2005 [10], FLASH has hosted a diverse range of experiments in Atomic, Molecular and Optical (AMO) physics over the past two years. The AMO user groups have come mainly from Germany but also from France, the Netherlands, Ireland, etc. Experiments have been performed on rare gas atoms and simple molecules with single EUV pulses and also with bichromatic fields using a two colour facility [11] which provides 800 nm/ 120fs/20 μJ pulses synchronised to the FEL. The objective of this brief Progress Report is to give a flavour of what's already possible with FLASH by recent salient results from some first experiments.

The paper is organised as follows. Section 2 comprises a brief introduction to FLASH and an outline of its current key performance indicators. Section 3 comprises an illustrative run through some of the experiments and salient results to date. 3 is broken into subsections on few photon processes, dilute targets and two colour photoionization studies. A brief perspective is given in Section 4.

2. *FLASH*: *F*ree-electron *LAS*er in *H*amburg

A vacuum-UV (VUV) free electron laser (FEL), operating at the then record short wavelength of 98 nm was reported by DESY in 2000 [12]. It soon reached GW peak power levels [13] and was used to perform what was ostensibly the first VUV multiphoton experiment on rare gas clusters [14]. FLASH recently achieved world record peak and average coherent power [15] at a wavelength of 13.7 nm while the 5th harmonic (~2.7 nm) reached into the 'water window' (figure 2).

As the fundamental wavelength of the emitted radiation scales as the inverse square of the electron energy, to drive the shortest design wavelength (6 nm) range, it was necessary to continuously increase the beam energy. By 2005 the VUV FEL at Hamburg had become the EUV FEL at Hamburg and was rechristened 'FLASH'. FLASH is based on a linear accelerator coupled to a series of long and precisely aligned undulators acting in concert (Figure 1).

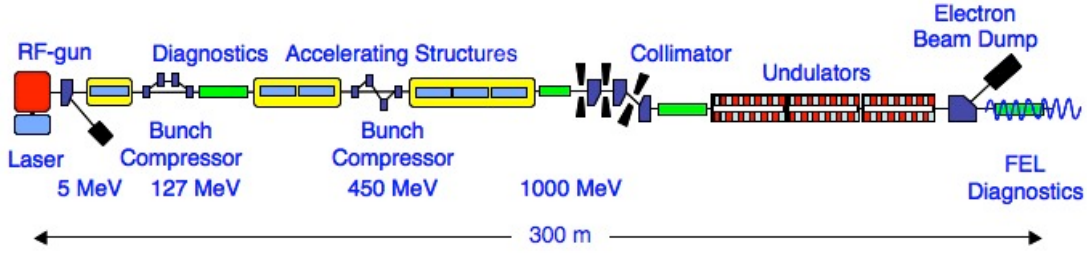

Figure 1. Layout of the EUV FEL 'FLASH' at DESY, Hamburg [10].

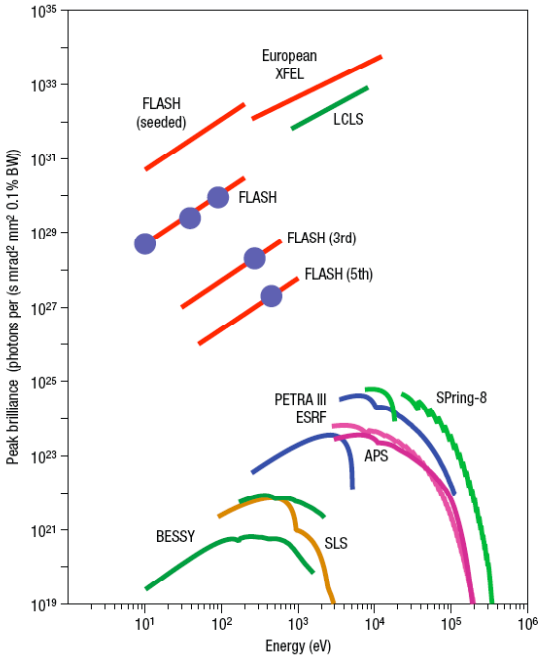

Figure 2. Measured FLASH brilliance [15]

Referring to figure 1, ultrashort charge packets (~1nC) are ejected from a photocathode, accerelerated and compressed to produce a bunchtrain of low emittance and low energy spread. Electron bunches are injected into the undulator chain, where they undergo a zig-zag motion and emit synchrotron radiation. Radiation emitted by deflected electrons moves faster than the electron bunches and interacts with electrons further up the beam, accelerating or decelerating them leading to a periodic bunching of the electrons that imposes a density modulation period equal to the undulator fundamental period. The imposition of a well defined spatial periodicity in the emitting bunches results in increasing coherence, collimation and an exponential growth in the intensity of the laser-like radiation field as the bunchtrain travels down the train of undulators. Since the FEL output grows out of spontaneous emission (effectively the photon noise), the process is referred to as *SASE– 'Self Amplified Spontaneous Emission'* [16].

3. Overview of Selected Atomic and Molecular Photoionization Experiments at FLASH

Photoionization pathways for atoms and molecules in laser fields depend on a combination of ionization potential, laser photon energy and laser intensity. Theoretical descriptions can be effected within a perturbative framework or may require a full solution of the time dependent Schrodinger equation depending whether the particular experiment resides within the multiphoton (MPI), tunnel (TI) or the related over the barrier (OBI) ionization regimes. A simple but instructive parameter (γ) that determines in which regime a particular experiment is likely to lie is due to Keldysh [17]:

$$\gamma = \sqrt{\frac{IP}{2U_p}} \text{ where } U_P = 9.3\times10^{-14} I\left(Wcm^{-2}\right)\lambda^2\left(\mu m\right) \quad eV$$

Here IP is the ionization potential and U_p is the so-called ponderomotive potential (the cycle averaged kinetic energy of a liberated electron in the laser field). As the laser wavelength moves into the EUV, the ponderomotive potential drops quadratically and the Keldysh parameter increases linearly. Hence EUV FELs will tend to favour multiphoton effects that are readily described by pertubation methods.

One way to look at this result is as follows. When the photon energy lies well above the ionization potential, the tunnel ionization rate will be less than the laser frequency and so the most favoured route to ionization is via multiple photon absorption. From another viewpoint, as the laser photon energy increases, fewer photons are needed to ionize the atoms and so the ionization probability becomes quite high at relatively low intensity (at least compared to visible/IR lasers). Taking the case of He, for a laser intensity of 10^{15} W/cm^2, $\gamma = 0.45$ for a Ti-Sapphire laser (tunnel ionization regime) while for FLASH operating at 8 nm, $\gamma \sim 45$ (very clearly the MPI regime).

The result is hugely important since it means that EUV FELs bring high intensity laser-matter interaction back into a space where few photon processes reign and accurate perturbative methods may be used to treat the problem. As one might expect, there has been a surge in theoretical activity since the late 1990s when firm design parameters for EUV/X-ray FELs started to appear. However, pioneering papers on intense VUV-atom interactions due to Lambropoulos and colleagues appeared as early as 1981 [18]. The recent papers on double ionization of He in intense EUV fields by

Nikolopoulos and Lambropoulos [19] and Ivanov and Kheifets [20] illustrate nicely the state of play in theory and make a good starting point for a journey through the recent history of activity.

Although FLASH has hosted a broad range of experiments including notably coherent imaging [21] and EUV laser-matter interactions [22], the focus of this section will be exclusively on AMO experiments. They break broadly into few-photon ionization, EUV laser induced fluorescence, molecular photodissociation, two colour above threshold ionization and pump-probe experiments. Salient, but by no means complete, results are shown in 3.1 to 3.3.

3.1. Few Photon Single and Multiple Ionization (FPMI) Experiments with the Rare Gas Atoms

Not unexpectedly this particular domain has received much attention given the promise for exciting new results which was brought into sharp relief by Phase 1 experiments [14]. Sorokin et al. [23] have performed systematic measurements on the photoion spectra of He and Ne under high irradiance (up to 10^{14} W.cm^{-2}). They chose two photon energies, namely 38.4 eV and 42.8 eV which lie just below and just above the Ne^+ ionization threshold respectively. They did so as a key objective was to separate sequential ionization (SI) from so called direct or non-sequential ionization (NSI) pathways in Ne. As the experimental apparatus could be moved with high precision through the optimum FLASH focus and the pulse energies could be measured accurately using a so-called Gas Monitor Detector (GMD) developed by this group [24], they were able to measure ionization yields for varying EUV intensity. Ion-yield ratios versus FLASH intensity are shown in figure 3.

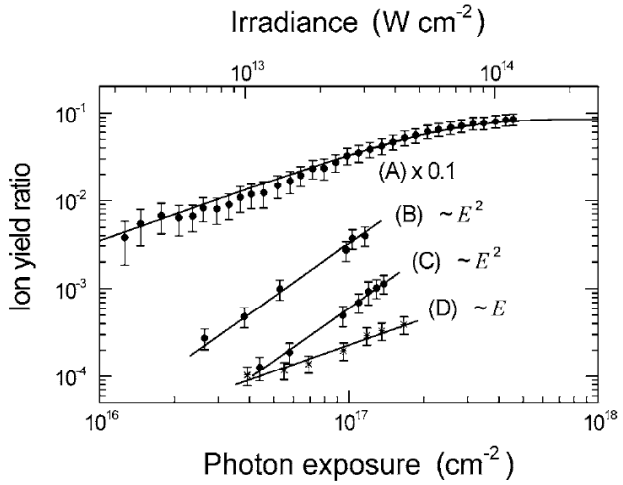

Figure 3. Ion yield ratios as a function of irradiance and photon density at 42.8 eV, (A - Ne^{2+}/Ne^+), (C - Ne^{3+}/Ne^{2+}), (D - He^{2+}/He^+) and for 38.4 eV (B - Ne^{2+}/Ne^+). A implies sequential two photon double ionization of Ne via Ne^+ as the production mechanism, B - one photon ionization of Ne followed by two photon single ionization of Ne^+, C (Ne^{3+}) requires the additional step of direct two photon single ionization of Ne^{2+}. D – two photon double ionization of He [23].

Using an analysis similar to that done earlier on N_2 ionization [25], Sorokin et al. were able to derive two photon ionization cross section for Ne, Ne^+ and Ne^{2+} (table 1 of Ref. 23). Their value of (1.6 ± 0.6) X 10^{-52} cm^{4}s at 42.8 eV for two photon double ionization of He is in good agreement with an earlier estimate of 1 x 10^{-52} cm^{4}s using focused HHG radiation at 41.8 eV [4] and recent theory [26].

Moshammer et al. [27] brought the Heidelberg Reaction Microscope [28] to FLASH to obtain impressive data and insights into the dynamics of few photon mutiple ionization of Ne and Ar. The microscope directs electrons and ions onto horizontally opposed position sensitive detectors. From the position and time of flight for each hit, the full momentum vectors of the photoionization products can be reconstructed. The FLASH output photon energy of 38 eV meant that sequential double ionization of Ne was forbidden.

The ion yield scaling with focused intensity (figure 4a) showed that non-sequential two photon double ionization (NSTPDI), evidence by the quadratic dependence of Ne^{2+} yield on intensity, dominates for I < 6 X 10^{12} W.cm^{-2}. For higher intensities a second process, referred to as three photon double ionization by the authors, is superimposed on NSTPDI. Here three photons are involved, one photon for the first step (Ne → Ne^+) followed by two photons for the second step (Ne^+ → Ne^{2+}). Recoil ion momentum distributions for Ne^{2+} are also shown in figure 4 for low (b) and high (c) FLASH intensity. For low intensity, the concentration of event around the zero momentum origin

leads to the conclusion that electrons are ejected into opposite directions (hemispheres) with roughly equal energies. This signal result stands in stark contrast to the one photon double ionization case where such a dynamical decay pattern is suppressed for equal energy sharing.

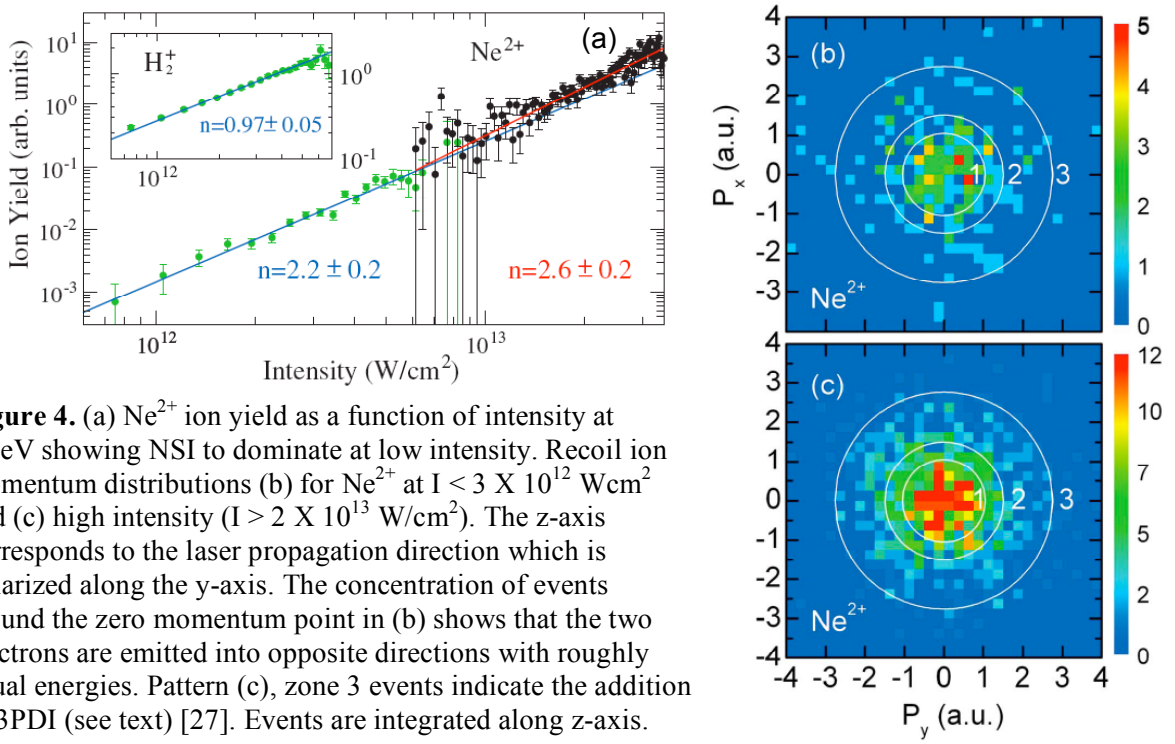

Figure 4. (a) Ne^{2+} ion yield as a function of intensity at 38 eV showing NSI to dominate at low intensity. Recoil ion momentum distributions (b) for Ne^{2+} at I < 3 X 10^{12} Wcm^2 and (c) high intensity (I > 2 X 10^{13} W/cm^2). The z-axis corresponds to the laser propagation direction which is polarized along the y-axis. The concentration of events around the zero momentum point in (b) shows that the two electrons are emitted into opposite directions with roughly equal energies. Pattern (c), zone 3 events indicate the addition of 3PDI (see text) [27]. Events are integrated along z-axis.

Becker et al. [29] have focused on their efforts on a systematic measurement campaign of the high resolution photoelectron spectroscopy on the rare gases and the small diatomic molecules. Results have yet to be published but an indicative result for atomic argon is shown in figure 5.

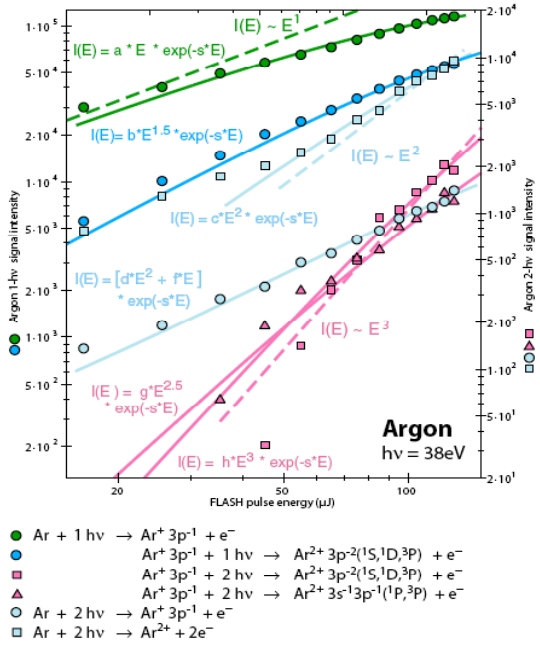

Figure 5. Argon ionization yield scaling with FLASH intensity [29].

The experimental system consisted of two vacuum chambers, one centred on the focus of FLASH with an array of time-of-flight electron spectrometers disposed at selected angles around the interaction point, the other located behind housing an ion spectrometer. Photoionization of argon at 38 eV exhibits the largest variety of multiphoton processes in this energy. Three distinct classes of intensity behaviour, involving anything from 1 – 3 photons are evident by inspection of the intensity scaling which yields exponents of 1, 2 or 3 respectively. the exception is the strong sequential 2 photon process deviates from the integer photon intensity scaling as it possesses an exponent of 1.5.

A resonant two photon absorption experiment on He has been performed by a University of Hamburg/DESY collaboration [30]. Working also at a photon energy of 38 eV they observed weak features at a kinetic energy of 24 – 26 eV which displayed a quadratic intensity dependence revealing a two photon origin, but significantly, only when FLASH was operated in multibunch mode. The authors concluded that this signal originated as follows - two photon excitation to doubly excited He states lying nearby the N = 5,6 thresholds of He^+ at ~77 eV followed by relaxation to lower lying doubly excited states. The photoelectron features at 24 - 26 eV are then produced in direct ionization by the next (~38 eV) micropulse in the FLASH pulse train. Föhlisch et al [31] have also recently reported an intriguing EUV field dependence of the valence photoelectron distribution of N_2.

The emphasis on especially two photon processes points up the need for reliable data on the harmonic content [32] in the FLASH output. Vrakking and co-workers [33] have been able to track and extract the 2nd (H2) and 3rd (H3) harmonic content for each micropulse in a long pulse train. Interestingly they find that while the third harmonic content is more or less constant as a function of micro pulse number, the second harmonic content increases by about 50% over the full macro pulse. Other new results yet to be published include velocity map images of molecular dissociation [33].

3.2. Molecular Photodissocation of HeH^+ and EUV LIF of Li-like Fe

Two recent results are chosen to demonstrate how this tour-de-force EUV FEL has made possible meaningful measurements on very dilute targets. One [34] concerns fragmentation of a molecular system of astrophysical significance (HeH^+) while the other illustrates how FLASH has brought a mature, but key, technique for the quantification salient QED terms in ions, namely resonance laser induced fluorescence (LIF), into the EUV for the first time [35]. Figure 6 shows a neutral species fragment image. The finding, that HeH^+ photodissociation occurs mainly normal to the polarization axis, showed the importance of effects so far left out of current calculations. High lying excitation pathways are proffered as a possible explanation, to be confirmed in more complete calculations.

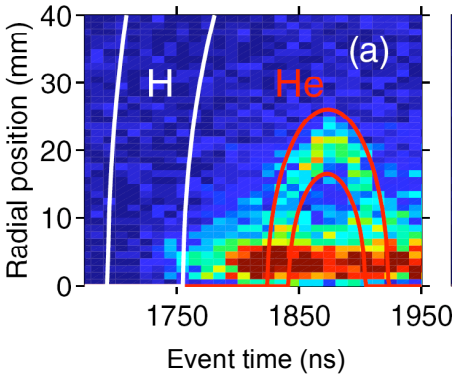

Figure 6. Photodissociation image of neutral fragments of HeH^+ [34]

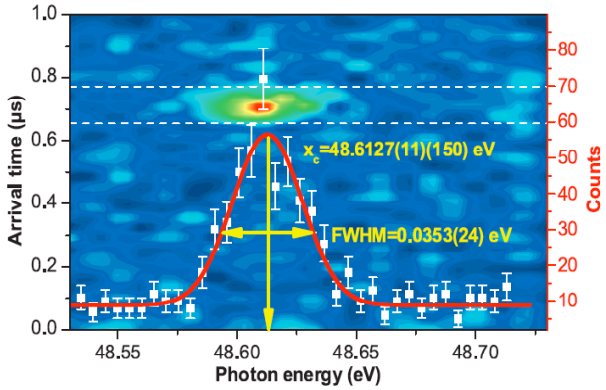

Figure 7. EUV LIF signal from of the 2s–2p transition of Fe^{23+} [35]

Figure 7 shows the very first LIF measurement on the resonance $2S_{1/2} \rightarrow 2P_{1/2,3/2}$ transition of Fe^{23+}, the highest energy, direct laser excited transition probed to date. The impressive measurement accuracy already achieved (22 ppm) meets the best conventional methods and will be significantly improved with the increased FLASH repetition rate in 2008. The relative accuracy already beats theoretical estimates for the dominant QED contributions (see figure 4 of Ref. 35) and points to a bright future for fundamental QED tests on highly charged ion systems at FLASH.

3.3. Two colour experiments with FLASH and an optical (800 nm) femtosecond system.

A femtosecond optical laser synchronized to FLASH and optical diagnostics kit was built within the frame of an EU funded collaboration involving DESY, MBI, BESSY, Orsay, Dublin and Lund. It can be used to perform both pump-probe and laser assisted photoionization (LAP) studies. In

LAP experiments, when the EUV and optical laser fields overlap in space and time, a photoelectron is born into the optical dressing field where it can gain (or lose) one or more quanta of energy. The resultant photoelectron spectrum displays sidebands adjacent to the main photoline separated by the optical photon energy. Sideband intensity depends on the EUV photon energy and dressing field strength and so can be used to monitor the relative time delay jitter between overlapping pulses.

We have developed an experiment [11] to determine FLASH harmonic content [32], study interference free sidebands [36] and determine timing jitter between EUV and FEL on a femtosecond timescale (250 fs rms) [37] based on LAP. The experimental layout, typical sideband spectra and timing jitter (< 600 fs FWHM) extracted are shown on figure 8.

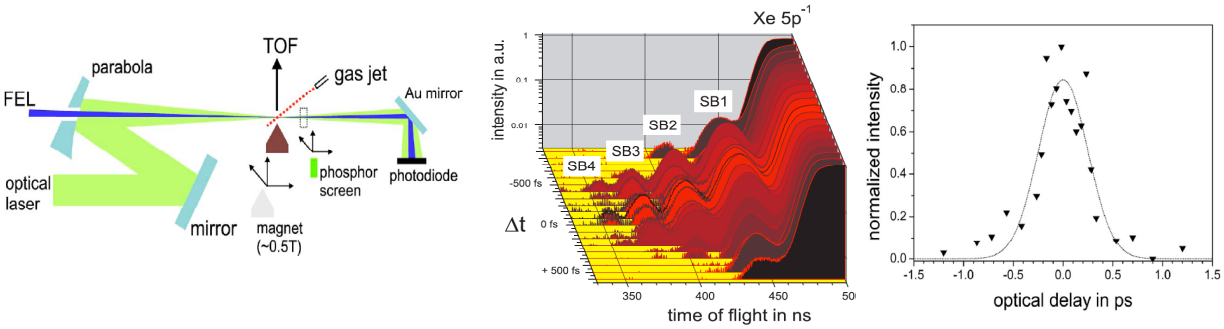

Figure 8. Experiment, typical spectra and timing data from laser assisted PES at FLASH [37].

We have also recently carried out experiments on resonant two photon inner shell excitation of Kr (3d – 4d) and excitation dynamics of H atoms, formed in dissociation of H_2 in a pump-probe experiment. Analysis and interpretation are ongoing. Further experiments along these lines are planned for 2008.

Drescher and collaborators have developed a photoelectron imaging system to directly measure and display pulse and timing jitter using the LAP technique – so-called time-space mapping [38]. The experimental scheme and cross-correlation trace are shown on figure 9.

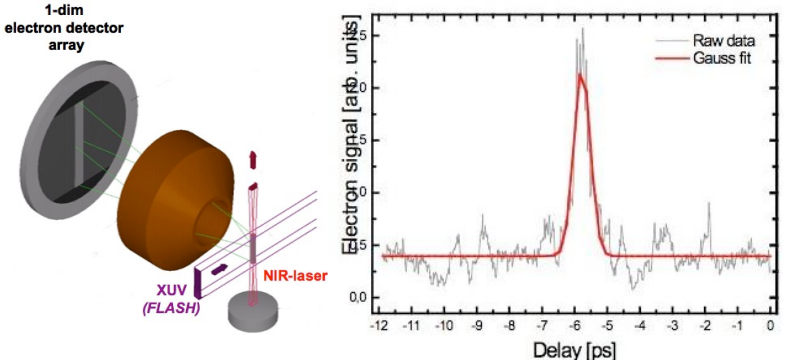

Figure 9. Experimental setup for laser assisted photoelectron imaging and EUV-optical pulse jitter measurement. Photoelectrons from a directed krypton gas jet produced at different points along the optical laser focal waist, corresponding to different delay times, are imaged to different points on a MCP [38]

Their value of 600 fs FWHM is in full agreement with our value [38]. A new cross correlation methodology, based on X-ray field modified surface reflectivity, which offers the promise a neat, simple and scalable on-line technique is under development by Drescher, Föhlisch, Wurth et al. [39].

4. Near Future Perspective

FLASH is a hugely significant milestone on the road to X-ray FELs operating at wavelengths comparable to the atomic unit of spatial scale and with pulse durations which will approach the atomic unit of time. Such XFELs will permit atomic structure and dynamics to be directly and simultaneously probed on intrinsic scales. Hence the atomic nature of systems ranging from cancer causing rogue molecules to complex nanostructures may be distinguished, extracted and possibly even controlled. All such future studies will rely on a continuing stream of innovative, critically executed and evaluated FLASH to XFEL photoionization experiments on simple prototypical atomic and molecular systems.

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