SASE FELs: Interactions with Atoms and Ions

E T Kennedy
National Centre for Plasma Science and Technology, School of Physical Sciences, Dublin City University, Dublin 9, Ireland

Abstract. Attainment of tunable laser-like radiation in the VUV and X-ray spectral regions is becoming a reality with the construction of single-pass Free Electron Lasers (FELs) based on the principle of Self-Amplified Spontaneous Emission (SASE). The paper introduces the development of short wavelength sources from an atomic physics perspective, describes the SASE FEL process and discusses examples from first user experiments at FLASH (Free Electron Laser at DESY (Hamburg). The perspective for further progress to shorter wavelength devices is outlined.

1. Introduction
Photoionisation is a fundamental process in nature. Experiments involving the interaction of ionizing photons with atoms and ions, the basic building blocks of matter, improve our basic quantum mechanical understanding of atomic physics processes and provide valuable data for modelling of laboratory, fusion and astrophysical plasmas. Lack of basic data is particularly acute for ions, despite their pre-eminence in nature. Progress in the study of the interactions of short wavelength photons depends critically on advances in atom/ion beams and detectors for the resulting ions, electrons or photons but most critically on the development of better light sources. The holy-grail is the attainment of short wavelength tunable laser light. Achieving laser action at short wavelengths through the conventional approach of population inversion is challenging however, as highly charged ions are necessary to provide the concomitant large inter-level spacing and level lifetimes scale very strongly with decreasing wavelength thus implying the need for extremely fast pumping mechanisms. These light sources are moreover limited to discrete wavelengths due to their reliance on bound electron levels and are of limited coherence and photon numbers per shot. An alternative approach and one that is now beginning to pay rich dividends is the development of laser quality radiation achieved using free electrons accelerated to relativistic speeds in linear accelerators.

2. Development of accelerator based light sources
In the development of accelerator based light sources over the last forty years or so, distinctive generations have been identified leading to ever-improving performance. The significance of storage rings as potential short wavelength light sources was highlighted for the atomic physics community through the pioneering work of Madden and Codling [1], who observed the landmark strong and weak asymmetric resonance series associated with the excitation of both 1s electrons of helium. The interpretation of the spectra required consideration of the correlated motions of the two electrons and led to the introduction of new correlation quantum numbers in the accompanying paper by Cooper, Fano and Prats [2].
The impressive experimental results and the new insight from the theoretical interpretation stimulated the photoionisation community and several experiments soon followed with other atomic species. This early work relied on parasitic use of storage rings originally built for particle physics; often termed first generation devices from the point of view of light source developments. Second generation machines specifically designed as light sources followed later and through the introduction of multiple photon beamlines began to cater for a wider user community quickly realizing the potential of these new light sources for a range of science investigations.

By introducing straight periodic permanent magnetic structures, known as undulators or wigglers, the electrons are forced to rapidly oscillate sideways, leading to more efficient emission of light. The systematic introduction of undulator-beamlines together with improvements in the quality of the circulating electron beams led to third generation sources such as ALS, APS, BESSY II, Elettra, ESRF, Max II, Spring 8, providing orders of magnitude improvement in performance compared with the earlier second generation facilities [3].

![Figure 1. Second versus third generation sources [5].](image)

Improvements in going from a second generation to a third generation machine can be substantial. Figure 1 shows an example taken from the interaction of short wavelength photons with free lithium atoms in the energy regime corresponding to the excitation of triply excited or hollow atom states. Observation of the electron energies associated with the decay patterns provides detailed information on the preferred decay routes. First photoelectron spectroscopy results were obtained at SuperACO on a bending magnet beamline (figure 1 upper panel). Later results on an undulator beamline on the same storage ring and at the third generation light source ALS at Berkeley California (middle and bottom panels, respectively) show substantially improved results [4,5]. Greatly improved count rates were obtained despite narrower monochromator settings for the excitation and narrower bandwidth for the electron spectrometer.

If the incident photon energies exceed 203 eV, single-photon triple-ionization of the lithium atom can be achieved. The measured cross section of only 6 Barns [6] implies that if triple-ionization experiments are to be achieved with coincidence detection of the outgoing electrons and recoiling nucleus then a much better light source is needed. A further example requiring better light source development is that of electron spectroscopy for ions. Because of the difficulty of obtaining ion beam densities comparable to atoms, the powerful electron spectroscopy technique has not generally been applicable to the study of the interaction of ionizing photons with ions, apart from the pioneering work of Bizau et al [7] using the very large cross section of about 2000 MBarns associated with the Ca$^+$ 3p $\rightarrow$ 3d excitation process. Further advances in light sources are therefore necessary if many desirable experiments are to be possible with atoms and ions, particularly those associated with very weak cross sections. What is needed is a short wavelength source with laser like qualities! High brightness and high flux would allow dilute targets such as highly charged ions to be investigated. Short pulse lengths would open up fast dynamic processes and coherent radiation would allow the full power of coherent imaging. Laser radiation at x-ray wavelengths matching the spatial scale of inter-atomic dimensions would open up structural biology and condensed matter fields.

3. Fourth generation sources

Recent advances in linear accelerators and developments in low-emittance electron bunches, now make possible the construction of Free Electron Lasers (FELs) based on self-amplified spontaneous
emission (SASE). These new fourth generation sources have the potential to provide uniquely intense, polarized, short-pulse radiation tunable throughout the VUV and X-ray regimes and exceeding synchrotron radiation and laser plasma sources by orders of magnitude in peak and average brilliance.

In a SASE FEL lasing occurs in a single pass of an ultra-relativistic electron bunch through a long undulator. The alternating magnetic field forces the electrons to oscillate sideways and emit electromagnetic radiation wave-packets, which are extremely strongly peaked in the forward direction in the laboratory frame of reference. In the early part of the undulator the radiation produced is the typical spontaneous radiation associated with the relatively short undulator insertion devices in third generation storage rings. If, however, the electron bunch is of high enough charge density and the electron beam and emitted electromagnetic beam paths overlap through a sufficiently long very precise undulator, then the emitted electromagnetic radiation interacts with the electron bunch leading to a density modulation (micro-bunching) which enhances the power and coherence of the radiation emitted by the bunch. The micro-bunching causes pile up of the electrons at intervals of one wavelength apart. The radiation wavepackets emitted by the microbunches are therefore in phase and add coherently, with the intensity now proportional to $N_{\text{elec}}^2$ as opposed to $N_{\text{elec}}$, leading to an increase in the photon pulse intensity by many orders of magnitude. This process of micro-bunching and self-amplification, known as the SASE process, leads to exponential growth of the radiation field [8]. As it is not constrained by the need for cavity mirrors, a SASE-FEL can, in principle, deliver very short wavelengths; the wavelength can be most readily tuned by varying the energy of the electrons from the accelerator.

4. The FLASH User facility at DESY Hamburg

The goal of the SASE FEL Phase 1 programme at DESY (Hamburg) was to provide experimental proof of operation of the SASE principle at short wavelengths and validation of the numerical codes which predicted the expected behaviour. DESY achieved a decisive milestone in February 2000 when the first coherent output was observed [9]. By 2001 the FEL had produced the first saturated output and achieved GW pulses in the femto-second regime [10]. The first scientific results were reported in [11] and involved the interaction of the VUV photons (at ~ 90nm) with xenon atoms and clusters (figure 2). Whereas Xe atoms only became singly ionised by the absorption of single photons, absorption in the clusters was strongly enhanced and high ion stages were observed (figure 2). The clusters absorbed many FEL photons and burst apart by Coulomb explosion. The behaviour was strikingly different to that which occurs with intense optical laser light.

The success of the Phase 1 machine has led to a Phase 2 (1 GeV) FEL development aimed at providing wavelengths extending down to 6 nm. This unique user facility, renamed as FLASH (Free Electron Laser at Hamburg), has enabled, during 2005 and 2006, laser operation at several wavelengths including 32 nm [12] and most recently in the 13 nm regime. Beam parameters of FLASH include virtually full transverse coherence, pulse duration < 100 fsec and ~ $10^{13}$ photons per pulse, making it a revolutionary new source. Several beamlines are being exploited by user groups carrying out first experiments. Many scientific areas including physics, chemistry, biology and materials science are benefiting.

![Figure 2](image-url)  
**Figure 2.** First results obtained from SASE FEL interactions with atoms and clusters [11]
4.1 Two-colour experiments on rare gas atoms at FLASH

As with the early synchrotrons, some of the first experiments with FLASH have concentrated on inert gases. A European collaborative effort has investigated the interaction of FLASH radiation with rare gas atoms, using Time of Flight (TOF) photoelectron spectroscopy and synchronized two-colour experiments [13].

Figure 4 shows the photoelectron lines produced from Xe by 38.5 eV FEL radiation. In addition to strong electron lines produced by the fundamental FEL wavelength, weaker lines due to second (77 eV) and third harmonics (115.5 eV) are also evident (shown on an enlarged scale in figure 4). Knowing the photoionisation cross sections at the different photon energies (figure 4) allowed an estimate of the harmonic contributions to be made. Both second and third harmonics were at the few tenths of a percent level compared with the fundamental [14]. The harmonics have also been detected with a grazing incidence spectrometer situated near the FEL entrance to the experimental hall [14] and a flat field spectrometer at the end of the beamline. The flat field spectrometer and photoelectron spectrometer results can provide a shot by shot measure of the wavelength content of the FEL beam and are a useful diagnostic for optimisation of the accelerator performance. Very recently harmonics have been observed to below the water window providing record performance in terms of coherent photons [15].

Figure 5 shows the setup for the two-colour experiments with collinear FEL and optical laser beams [13]. The TOF photoelectron spectrum shows sidebands (figure 6) adjacent to the strong main lines, when FEL induced photoionization takes place in the presence of the intense dressing optical laser field, (due to the exchange of photons from the optical laser field). First experiments utilised a 12 psec
long optical laser pulse in order to facilitate temporal overlap of the pulse [13]. Later experiments used 120 fsec laser pulses [16]. The sidebands can be seen to only exist within a scanning interval of about 600 fsec FWHM. This provides an upper limit to the jitter between the two very different FEL and optical laser systems, enables setting the zero time delay, and proves the feasibility of the facility for future two-photon experiments.

4.2 Non-linear processes at FLASH

An early objective of several experimental groups working at FLASH has been the unambiguous observation of nonlinearities in the interaction of the intense FEL light with atoms. Angle resolved photoelectron spectroscopy can provide additional information on the interaction of the intense FEL laser light with atoms. By studying the angular distribution of the electron emission as a function of the FEL pulse intensity at 76 eV (twice the fundamental photon energy of 38 eV) evidence for nonlinear processes associated with 2\omega effects was highlighted [17].

Quantitative absolute measurements of the output photon pulses are very important in terms of the FEL diagnostics and development. Radiation hard gas ionisation detectors have been developed [18] which provide single shot measurements to users of the individual absolute pulse energies (and therefore photons per pulse). This is extremely important in view of the shot-to-shot fluctuations due to the SASE process starting from noise [19]. The effect of the unprecedented number of photons per pulse of the FEL is strikingly illustrated through the nonlinear dependence of ion signals as a function of the photons per pulse. Figure 7 shows the sub-linear growth of the ion signal recorded with neon with incident photon number due to vanishing targets [20]. Essentially the early parts of the pulse deplete the focal volume of neon atoms due to ionisation, so the latter parts of the pulse have fewer neutral atoms to generate Ne\textsuperscript{+} ions. The saturation effect depends on \( \sigma N_{ph}/A \) [20] and because \( N_{ph} \) can be measured and \( \sigma \) is known, the cross sectional area of the FEL beam can be deduced. By scanning the ionisation chamber along the FEL beam the focus position and focal spot size can be determined in a non-destructive fashion.

The unprecedented numbers of photons per shot has also been evidenced through the single shot scattering pattern recording from a single Ar cluster of radius 32nm. Despite the complete destruction of the cluster by the pulse the 2-D scattering pattern agreed very well with the calculated pattern. The extremely short duration of the pulse essentially means that the scattering pattern is recorded before the cluster has time to blow apart [21].

Figure 6. Two-colour sidebands as function of the inter-laser delay [16]

Figure 7. Sub-linear growth of Ne\textsuperscript{+} ions with photons per pulse [20]
4.3 Interaction of FLASH with highly dilute samples

A key part of the development of the FLASH facility has been the combination of the FEL with specifically designed atom/ion/molecule sources and detector systems aimed at enabling single- and multi-photon differential spectroscopic photoionisation studies of highly dilute samples. The group from MPI-K at Heidelberg and their collaborators are exploiting an EBIT to supply highly charged target ions, Reaction Microscopes to enable coincidence measurements of few-photon few-electron interactions and Molecular Ionisation sources to provide molecular ions, particularly of astrophysical interest [22]. The facilities will uniquely allow the study of dilute atom, ion and molecule systems with the SASE FEL.

The EBIT can provide a very wide range of highly charged ions. Initial experiments on precision spectroscopy of lithium-like systems are aimed at providing improved measurements of the QED contribution to the level spacings. The FLASH monochromator beamline was used to provide narrowband FEL radiation to excite the 2s1/2 to 2p1/2 transition in Fe23+, with observation of the resulting fluorescence radiation confirming the excitation [22]. In future beamtimes it is anticipated that experiments will provide also differential spectroscopic experimental data for the first time on many ions.

The advent of SASE FELs has stimulated many calculations of the effects of short wavelength high intensity pulses with few-electron atomic and ionic species [23-35]. While single-photon ionisation of helium has been previously experimentally studied in great detail, there is wide interest in the potential of FLASH for studying few-photon few-electron ionisation processes in helium and lithium, the fundamental three-body and four-body atomic systems. A magneto-optical trap has been installed to provide cooled lithium atoms to uniquely enable differential spectroscopic studies of e.g. single-, double-, triple-ionisation of the three-electron system, despite the very low cross sections. These experiments will serve as a benchmark for theoretical calculations.

During the initial beam-times the wavelength and pulse repetition rate of the FEL were not yet appropriate for studying these processes and multi-photon photoionization effects were sought in neon and argon. Figure 8 shows an ion time of flight spectrum for argon, recorded at a photon energy of 29 eV, which shows sharp peaks of multiply charged argon ions superimposed on broad contributions from the background gas [22]. By studying the individual argon ion yields as functions of the FEL intensity, evidence for instantaneous non-sequential absorption of up to five photons was deduced. During the same beamtime the ionisation and dissociation dynamics of molecular hydrogen (D2) were investigated, again indicating simultaneous multi-photon absorption effects.

The above examples show that the first year of operation has enabled various teams of researchers to begin investigating the unique possibilities associated with the FLASH facility. It is anticipated that with ongoing developments in the FEL and optimisation of the beam-line experimental systems, our knowledge and understanding of intense short wavelength multi-photon processes in atoms and ions will be greatly improved over the coming years.
5. Perspectives for shorter wavelengths

In 2007 FLASH will be upgraded to emit tunable radiation down to 6 nm in the fundamental. Many other synchrotron facilities around the world are in the process of designing and building FELs for short wavelength operation. Several aim at the VUV and soft x-ray regions. Moving to even shorter wavelengths places extreme demands on the qualities of the accelerators and undulators required. Nevertheless, projects are underway [36,37] to develop FELs aimed at producing photon beams stretching through the water window (2.4 ≤ λ ≤ 4.4 nm; important in many biological applications), down to wavelengths of the order of ~ 0.1/0.2 nm – the natural dimension of atoms, molecules and material inter-atomic spacings. Such X-Ray FEL lasers would constitute scientific investigative tools of dramatically new capability. The Linear Coherent Light Source (LCLS) FEL at Stanford [36] is to become operational in 2009 while DESY plan XFEL, a European user facility [37], aiming at completion in 2012. The XFEL Technical Design Report [37] completed in July 2006 describes the exciting science that will become possible.

The revolutionary nature of these sources is illustrated in figure 9, which shows the peak brilliance of FLASH compared to existing synchrotron facilities and indicates the expected performance of the Stanford and DESY XFEL devices. The figure shows FLASH with and without seeding. The latter involves removal of the random spiking in the temporal and spectral domains of the FEL pulse and will further increase the range of scientific applications, for example those requiring high spectral resolution. Conceptual designs involve seeding the FEL amplifier with a sufficiently narrow band of radiation at a power level well above the effective power of the shot noise in the electron beam, to improve the longitudinal coherence of the output pulse [38]. The narrow-band seeding pulse initiates the SASE process developing a homogenous microbunching potentially resulting in the final emission of a fully coherent FEL beam, with all the power in an energy window of ∆E/E < 10⁻⁴. The spectral brilliance should be greatly increased and intensity fluctuations reduced to a small level.

The success of FLASH and the prospects of SASE FELs extending down to x-ray wavelengths [39] introduce a new era in the study of photon-atom/ion interactions at short wavelengths. The availability of intense tunable laser radiation sources at ionising wavelengths is expected to pay rich dividends in terms of science and technology applications and will undoubtedly lead to many new and unforeseen developments in our understanding of fundamental photon-atom/ion interactions.

Figure 9. Peak brilliance for different sources [37]
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