Extended abstract

Coulomb acceleration of protons with a free-electron laser
Prospects for nuclear fusion

X-ray free electron lasers (XFELs) are a new kind of light source, capable of producing super-brilliant, ultra-short X-ray pulses with more than 10 orders of magnitude higher peak brilliance than pulses from synchrotron sources. This extremely high peak brilliance allows us to deposit huge amounts of energies into the sample over a very short time, and will revolutionize many areas of physics, chemistry and biology in applications like the imaging of single biomolecules, or the creation of highly ionized dense states of matter.

Molecular dynamics simulations, performed in Uppsala in the context of imaging biomolecules, have predicted the ejection of protons with unexpectedly high energies, of the order needed for fusion reactions. These protons are expelled from a nanometer-sized sample in a Coulomb explosion, caused by interaction with an XFEL beam. This motivated experiments at the FLASH facility, in Hamburg to study the interaction of the XFEL beam with both solid and clustered gas samples, and to look for signatures of highly energetic protons or deuterons, produced during the Coulomb explosion of samples rich in hydrogen or deuterium. The energies detected, in the solid case, were high enough to support the hypothesis of fusion. These results were analyzed and compared to predictions from a plasma code.

Improvements are still needed, mainly in an experimental point of view, and some of them are suggested here; however the way is opened for a very exciting new area of research related to XFELs.

X-ray free electron laser (XFEL), fusion, Coulomb Explosion, TOF (time-of-flight), plasma simulations, Molecular Dynamics

Introduction

X-ray free electron lasers (XFELs) can reach much higher beam intensities at much shorter wavelengths than usual lasers, and allow huge amounts of energy to be deposited into a sample during a few femtoseconds. FLASH (“Freie Elektronen Laser in Hamburg”), located at DESY (“Deutsches Elektronen Synchrotron”), in Hamburg, is the only operational XFEL reaching into the soft x-ray regime [1].

Free electron lasers (XFEL) differ from usual lasers by the lasing principle they use. Instead of relying on atomic or molecular bond states to produce coherent radiation they use electrons, accelerated to relativistic speeds, which radiate when going through an undulator,
with carefully chosen characteristics. FLASH is a high gain XFEL, based on the “Self Amplified Spontaneous (or Stimulated) Emission” (SASE) principle.

![2 fs (FWHM) FEL PULSE](image1)

**Figure 1:** MD simulation of radiation-induced Coulomb explosion of a small protein molecule (lysozyme). White balls: H, Gray: C, Blue: N, Red: O, Yellow: S. Integrated X-ray intensity: $3 \times 10^{12} (12\text{keV})$ photons/100 nm diameter spot. The picture shows the protein exposed to a 2 fs FWHM X-ray pulse, and its disintegration followed in time. The atomic positions in the first structures are practically identical at this pulse length due to an inertial delay in the explosion. Hydrogen ions and highly ionized sulphurs are the first to escape the immediate vicinity of the protein. The encircled stage is shown in detail in Figure 2.

![SLOW PROTONS](image2)

**Figure 2:** High-energy protons leaving the molecule very early during the explosion (encircled stage from Figure 1). The energy of the fast escaping protons strongly depends on their original chemical environment. In the case of lysozyme, two protons shoot out at very high energies already at the beginning of all simulations (they are outside the picture frame in Figure 1). These protons are attached to two specific sulphur atoms in the native protein, and leave in the same direction in a reproducible manner.

The use of XFELs has been proposed to achieve single-shot high-resolution images of molecules [2], the final goal being to image macromolecules and living cells with atomic resolution. Molecular dynamics simulations of the Coulomb explosion of a molecule [3, 4], presented in Figure 1, were made in the context of the imaging experiments. A sample is hit by a 2 fs XFEL beam and loses its electrons. It then explodes, given the repulsive Coulomb interaction between the remaining positive ions, but only after the end of the pulse, i.e. an image taken during the illumination of the sample shows it unchanged. Figure 2 corresponds to the encircled stage in Figure 1, and shows the unexpected acceleration of light ions, which follows the Coulomb explosion.

This observation set the possibility of accelerating protons or deuterons to KeV energies, in a few femtoseconds, by means of the interaction of solid or clustered samples with an XFEL beam. Deuterons accelerated to such energies are in good conditions to
undergo DD fusion reactions [3]. On a longer term, the aim will be to detect signatures of the following reaction [5]:

\[ D + D \rightarrow ^3He + n, \]  

\[ (1) \]

in the form of 2.45 MeV neutrons and 0.82 MeV alpha particles. The best candidates are neutrons, neutral particles which can easily escape the interaction region, and which leave with an energy characteristic of the reaction.

Intercluster fusion studies have been made, both theoretically [6] and experimentally [6, 7] and so have intracluster fusion studies [8], on a theoretical level, based on the interaction of ultra-intense optical/IR lasers with matter. However, in this case, the shift to x-ray frequencies, keeping a high intensity and a pulse duration of a few femtoseconds, makes the situation totally distinct, and opens the door to challenging new processes.

**Experiments: results and analysis**

Experiments on gas clusters were realized in March 2007, at DESY. Two gases were used, for forming clusters: argon, in a first step, since rare gases behavior is well known; and methane, the object under study, in a second step. The chamber, the cluster generator, and the ion TOF detector were provided by the cluster group at the Technischen Universität Berlin. In our case, no energy discrimination was possible for the ions. Beam intensity measurements were made by means of the GMD located before the XFEL shutter of the experimental hall. During these experiments, the XFEL was operating at a 13.5 nm wavelength (92 eV photons). The pulse energy was on average 20 µJ, but could go up to 40 µJ, and its duration was of about 10 fs; the focusing achieved corresponded to an area of around 5 µm² (FWHM). The pulse intensity was therefore of about 4.10¹⁶ W cm⁻² s⁻¹. In order to study the influence of the gas backing pressure – the pressure in the gas reservoir, before the cluster generating nozzle – in the cluster formation, it was set to both 300 mbar and 7 bar, in the case of Ar, and both 1 bar and 10 bar, in the case of methane. The XFEL beam was operating in multibunch mode, with a shooting frequency of 100 Hz.
Figure 3: Intensity dependence of the $H^+$ ions energy, for a 10 bar backing pressure.

Gas experiments results do not show any visible light ions energy increase due to the
presence of clusters or to an increase in the beam intensity (Figure 3), which would be
expected given the results obtained for solids, and those presented for clusters in the
literature [9]. However, the absence of fast ions in the spectra could be explained by a poor
adaptation of the TOF to energies of the order of 1 keV or higher, or by its geometry, namely
the size of the slit the ions have to go through to reach the MCP detector. Gas and gas
clusters experiments are part of an ongoing project and will be pursued in the next run of
FLASH.

Experiments on solids were performed in September 2006, at FLASH. The samples
used consisted of niobium rods, doped with different amounts of hydrogen and deuterium, a
tantalum plate doped with hydrogen, and a PMMA (Polymethyl methacrylate, C₆H₆O₂) plate.
Both the chamber and the detector used for these experiments belong to Ryszard Sobierajski’s team, in Warsaw. The detector consisted of an ion time-of-flight detector (TOF).
In our case, no energy discrimination was possible for the ions reaching the TOF’s MCP
detector. A measure of the beam intensity could be obtained through the output of the GMD
(Gas Monitor Detector), located just before the XFEL shutter, at the entrance of the
experimental hall. As for the characteristics of the XFEL, during the experiment itself, a
wavelength of 21 nm, corresponding to 51.9 eV photons, was used. The beam energy
varied between 1 and 20 $\mu$J, and the pulse length between 10 and 20 fs. The focal spot
had a 20 $\mu$m diameter (FWHM), i.e. a 314 $\mu$m² area. As a consequence, the beam
intensity attained was at the most $10^{14}$ W cm⁻² s⁻¹. The experiments were performed in
single bunch mode, two consecutive shots being about 2 s apart from each other. Only some
of the available samples were used, namely Nb, NbD, NbH, NbDH (relative concentration of 1, for H and D), TaH and PMMA.

A typical TOF output is presented in Figure 4, for PMMA.

![TOF spectra of PMMA, 1st spot, 23d shot](image)

**Figure 4: TOF spectra for PMMA.**

During the interaction of the XFEL beam with solid samples, H\(^+\), D\(^+\), and what was assumed to be D\(_2\)^+ ions were detected, with energies of up to 1\(\text{KeV}\), as seen in Figure 5. It was noticed that there are three different contributions to the H\(^+\) ions yield, reflected in the spectra as a split hydrogen peak.

![Hydrogen ions energy](image)

**Figure 5: Energy distribution for H\(^+\) ions (from three different origins), fitted by three gaussians.**

Three different contributions can thus be identified, for the H\(^+\) yield, which might account for different chemical origins or ejection mechanisms.
Figure 6: Intensity dependence of the time-of-flight for the H+, D+ and D2+ ions in NbDH. An increase in the kinetics energy (a decrease in the tof) of the ions follows the increase in beam intensity. Figure 6 is built on data coming from many shots. It therefore proves the consistency of the collected data among each other.

Furthermore, the energy of the ions was seen to increase for increasing beam intensity, as seen in Figure 6. The energy increase occurs differently for different ions, and also for each kind of H+ ions. The beam intensity increase also contributes to a larger yield of higher charged ion species.

Simulations were made using the code Cretin, developed at LLNL (Lawrence Livermore National Laboratory), by Howard Scott [10]. Cretin is a radiation transfer plasma code, which combines atomic kinetics and radiation transfer, and which can describe the behavior of a non-LTE (non Local Thermodynamic Equilibrium) plasma. In each step it updates the state of the system, based on initial input values of the density and temperature of the plasma, and on the energy and intensity of the incident beam. The atomic populations can be calculated either in a steady-state or a time-dependent mode. This code can run in 0D to 3D.

Cretin corresponds to a static description, which does not include a hydrodynamic expansion or a charge induced repulsion (leading to Coulomb explosion). In our case, it was used to calculate the heating of the material – more specifically the electron temperature – before the motion occurs. During this time, the thermal energy of the ions can be calculated from the electron-ion coupling coefficient.
This contribution is found to be smaller than the values observed experimentally. In our experiments, the faster hydrogen ions have kinetic energies of up to $1\, keV$ (Figure 5), whereas the simulations report ions of $0.1\, keV$ at the most (Figure 7). Only 10% of the energy of the ions can thus be explained by thermal heating in the initial phase of the interaction. The remaining acceleration must be explained by other mechanisms, such as Coulomb acceleration, in which the ions are further accelerated by charge repulsion from the positively charged core of the sample.

It is difficult to have a good estimate of the contribution of the Coulomb explosion, through simulations. Cretin does not include this kind of mechanism, and MD simulations, performed for hard x-rays interactions, might not be entirely accurate in this energy range. Other simulation methods, namely particle-in-cell [8], would be required, in order to have a good estimate of the Coulomb acceleration contribution to the final energy of the ions. This would confirm whether this is the only effect that should be considered, or if other acceleration mechanisms should be taken into account – such as space charge effects, as described by Mora in [11] – which should be relevant in ionized materials.

Conclusions and outlook

This study represents the beginning of a larger project, whose aim is to analyze and define conditions for achieving fusion through the interaction of an XFEL beam with both solid samples and gas clusters. The first experiments performed at FLASH in this context have yielded very exciting results, in respect to solid samples, through the observation of $H^+$ – and some $D^+$ – ions with energies up to $1\, keV$.

We have observed considerable proton emissions in which the energy of the emitted protons is very high (many hundreds of $eV$ to $KeV$), namely higher than expected from simulations made with an atomic physics code, Cretin. Furthermore, theoretical models
grounded on Molecular Dynamics predict different energies for protons coming from different chemical environments (including protons originating from adsorbed waters or hydrogens on the surface). We, therefore, expected to see split or multiple proton peaks in the TOF signals, and they were indeed present in the results. The models also predict that the energy of protons coming from different chemical environments will change differently with the FEL pulse energy. We have observed this and have seen a larger proportion of faster protons at higher beam intensities, in solid samples. Moreover, we expected to see more than one mechanism behind proton ejection, and the gaussians fitted to the data from solid samples confirmed such an expectation. Finally, methane cluster studies are an ongoing project, where experimental details need to be worked out.

These promising results led to the allocation of dedicated beamtime for this project during the next run at FLASH, starting in the end of 2007, and to the development of collaborations, necessary to cover the broadness of the experiments and of the analysis required. Several improvements are necessary, namely regarding the solid and gas samples – chosen materials, amount of hydrogen and deuterium, polishing of the surface, in the case of the solids –, the available diagnostics – type of detectors and their adaptation to the experiment –, and the overall characteristics of the beam – focusing, wavelength, stability –, some of which can, and will, be addressed in the next experiments. Apart from these improvements in the experiments, one must both develop tools for analyzing the data, and compare the outcomes results with simulation models (based on Molecular Dynamics or particle-in-cell) that are better adapted to this particular matter. Eventually, XFELs will start operating at hard x-rays wavelengths, opening new possibilities for all kinds of research, among which fusion related projects.

References

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