Free-Electron Laser Projects at DESY-Examples of Science at FLASH

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Abstract Free-electron lasers will allow direct investigations of matter with atomic resolution in space and time, and will open windows into unknown territories in science and technology. They nicely complement research at synchrotron radiation storage rings as well as research at so called table-top lasers. Several FELs for hard X-rays are under construction, one at SLAC in California, one at SPring-8 in Japan, followed by the European XFEL Facility in Hamburg. FLASH in Hamburg is the first FEL user facility in the spectral range of the EUV and in operation since summer 2005. It is a prototype for hard X-ray facilities both with respect to accelerators and to teaching the potential users to make the best out of it. The success of FLASH raised the international attention and interest in this new class of research tools.

1. Introduction

For more than 110 years X-rays play a key role in medicine and materials science, as well as in fundamental research. Enormous progress has been made since the first use of synchrotron radiation in the nineteen-sixties mainly due to the improvements of electron storage ring facilities leading to a gain of 3 orders of magnitude in brilliance every 10 years. Beamline technology and X-ray optics followed this development. The electron storage-ring technology is currently getting close to its theoretical performance limit and in order to further promote the field nevertheless, most modern synchrotron radiation facilities concentrate on improving undulators, optics, sample environment and on developing soft ware tools, data handling procedures and detectors. The development of the latter still seems to be slow. Over the years competent user communities were built up all over the world doing outstanding science very efficiently. With improved X-ray sources new fields of application showed up and today many laboratories aim for building up outstations on their campus which are focused on thematic research and thereby improving strongly the research opportunities of the widespread general user communities.

Today, in most cases we study equilibrium states of matter and time resolved X-ray scattering is just in its infancy, although we all agree about the importance of observing physical or biological systems ''at work'', getting from static pictures to movies. This is partly due to the fact that the individual X-ray flashes from storage rings are too weak and with 50 to 100 picoseconds duration they are too long compared to nature's time scale, which reaches to femtoseconds, and even attoseconds when it comes to electron dynamics. Storage rings cannot produce the type of radiation needed and therefore people look for new types of X-ray sources. Tremendous progress is observed with high power optical lasers but things get very difficult when one aims for intense subpicosecond pulses of hard X-rays which are needed to get the spatial resolution on the atomic scale. The development of powerful low emittance electron guns in the nineteen-nineties opened the possibility to realize accelerator driven X-ray sources which use linear accelerators and strongly exceed the performance limits of the storage ring technology.

Conceptually the next step beyond the storage ring is the so called energy recovery linac (ERL) originally proposed by M. Tigner in 1965 in the context of linear colliders (**Fig. 1**). Short, low emittance electron bunches

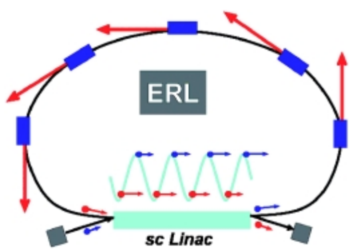

Fig. 1 Principles of an X-ray source with an Energy Recovery Linac operated with superconducting radio frequency.

are accelerated in a linear accelerator with superconducting RF up to 6 GeV as an example, circulated through a ring with a larger number of undulators, enter the linac a second time where they get decelerated to energies ideally below 10 MeV and guided into a beam dump. Such a facility avoids the equilibration in a storage ring, preserves transverse emittance and short pulse duration and recovers most of the energy in the electron beam due to the deceleration before entering the dump. In comparison to modern storage ring facilities ERLs provide higher spectral brilliance and flux, shorter X-ray pulses and smaller X-ray source size for nanometer size beams. The ERL principle has been demonstrated at the Jefferson Laboratory in the US where a free-electron laser for the spectral range of the IR is operated in an energy recovery scheme for many years. There are still a number of crucial technical issues to be understood and vigorous R&D programs for hard X-ray ERLs are pursued at Cornell University, USA, and also at KEK in Japan.

While ERLs may be considered as a natural next step beyond storage rings, one expects a revolution in X-ray science from single pass free-electron lasers providing radiation from soft to hard X-rays. We expect gains up to 10 orders of magnitude in peak and about 4 orders of magnitude in average brilliance. The beam is transversely coherent and the pulse duration is between 10 and 100 femtoseconds. Today we get at ~13 nm in a flash of 10 femtoseconds duration as many photons as we obtain in the best storage ring facilities per second. The first X-ray FELs for hard X-rays will work according to the principle of Self Amplified Spontaneous Emission (SASE) (**Fig. 2**). Electron bunches of high charge and low emittance are produced in a gun, e.g. via photo emission, accelerated to the several GeV level and guided through a long undulator. Because of the lateral velocity component of the electrons the photon field produced by the electron bunches in the previous part of the undulator can couple to the charge in the electron bunch and produce a modulation of the charge distribution with a period equal to the wavelength of the undulator radiation. This periodic distribution of free electrons is then brought to emit laser like X-rays via stimulated emission triggered by the already existing photon field. In order to drive this process into saturation the undulator may become very long, e.g. longer than 100 m.

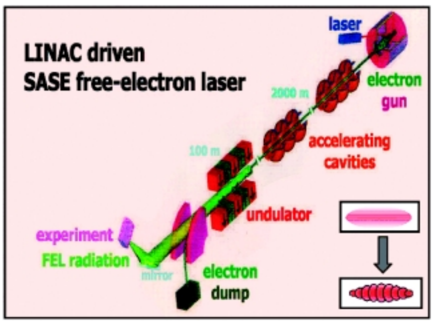

Fig. 2 Main components of a SASE free-electron laser where the originally homogeneously distributed electron charge in a bunch is transformed in a modulated structure with a period equal to the wavelength of the undulator X-rays.

2. The European XFEL Facility in Hamburg

The facility starts with the electron injector on the DESY site which allows the usage of very substantial existing infrastructure. The linear accelerator uses superconducting RF technology as developed by the international TESLA collaboration since the beginning of the nineteen-nineties. The repetition rate of ~1 msec long bunch trains, which contain up to 3000 electron bunches of ~100 fsec duration, is 10 Hz. The electron bunches are accelerated to an energy of 17.5 GeV and then distributed on 5 radiators, 3 of them are FELs, the other 2 produce spontaneous radiation with energies reaching several hundreds of keV. All tunnels as well as the injector building and the experimental hall are underground (**Fig. 3**).

When operated at an electron energy of 17.5 GeV the spectral range of the FEL radiation ranges from 0.1 to 1.6 nm, when operated at 10 GeV the range extends to 4.9 nm for the tunable helical undulator (**Fig. 4**). In addition two spontaneous undulators U1 and U2 provide short pulse X-rays in a wavelength spectrum ranging to way below 0.01 nm. 10 instrumental sta-

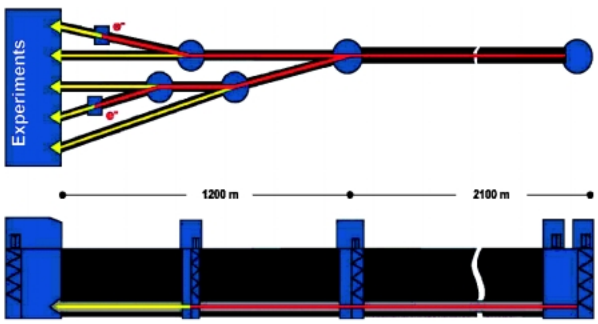

Fig. 3 Schematic Layout of the underground tunnel system of the European XFEL Facility in Hamburg.

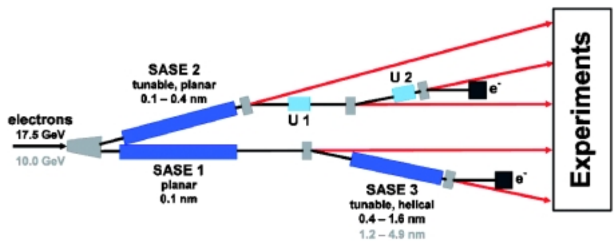

Fig. 4 Insertion devices at the European XFEL Facility and wavelength range of FEL radiation produced when operating the LINAC with 17.5 GeV or 10 GeV, respectively.

tions are included in the baseline design of the facility which is described in the Technical Design Report published in summer 2006 and available under http://xfel.desy.de/tdr/index_eng.html. The total cost of the facility at year 2005 prices is 1082 M€ including preparation and commissioning. The construction costs amount to 986.4 M€. Recently it has been decided to realize the facility in two steps, the construction costs of phase 1 with only 3 insertion devices, but still covering the whole wavelength range, amount to 850 M€. Before embarking on such a big enterprise the SASE principle had to be verified and the many novel components of the accelerator including undulators needed to be tested. Therefore the TESLA Test Facility (TTF) was built at DESY out of which FLASH grew, the Free-Electron Laser in Hamburg serving the spectral range from about 80 to 6 nm in the fundamental.

3. FLASH, the EUV free-electron laser at DESY

The facility consists of a laser driven photocathode electron gun followed by a combination of accelerator modules, bunch compressors and a collimator protecting the 6 undulators, each 4.5 m long, against dark currents (**Fig. 5**). An electron bypass bridging the undulators facilitates accelerator studies. The undulators are fixed gap devices in standard technology but with much more demanding specifications on the magnetic field and the overall stability. The shortest wavelength reached so far with the FEL in saturation is 13.4 nm, and we obtain about 10^{12} photons in flashes of 10 fsec duration. FLASH is currently the most powerful laser in the spectral range of the EUV. Right now the accelerator is extended to reach the nominal electron energy of 1 GeV, and for the fall this year it is hoped to achieve lasing at 6.5 nm in the fundamental. All along the trajectory different types of electron beam diagnostics are installed including tools to measure the longitudinal distribution of charge in the electron bunches. The performance of FLASH fully agrees in all aspects with theoretical calculations which are also the basis for the design of the European XFEL Facility (*W. Ackermann et al.*, NATURE *Photonics* **1**, 336 (2007)) (**Fig. 6**). The output beam also contains a significant contribution from odd harmonics of approximately 0.6% and 0.03% for the 3^{rd} (4.6 nm) and the 5^{th} (2.75 nm) harmonics, respectively, when comparing with the fundamental. At 2.75 nanometers the 5^{th} harmonic of the radiation enables FLASH to reach deep

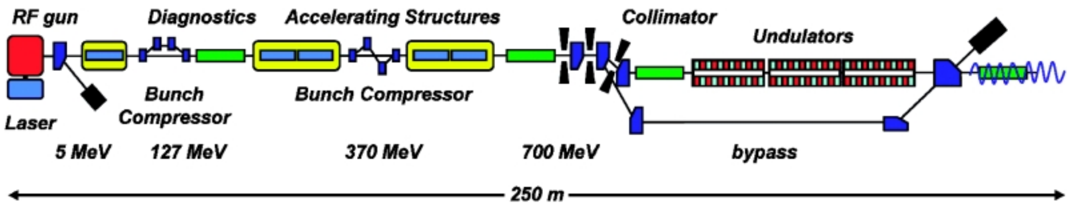

Fig. 5 Schematic layout of the accelerator part of FLASH.

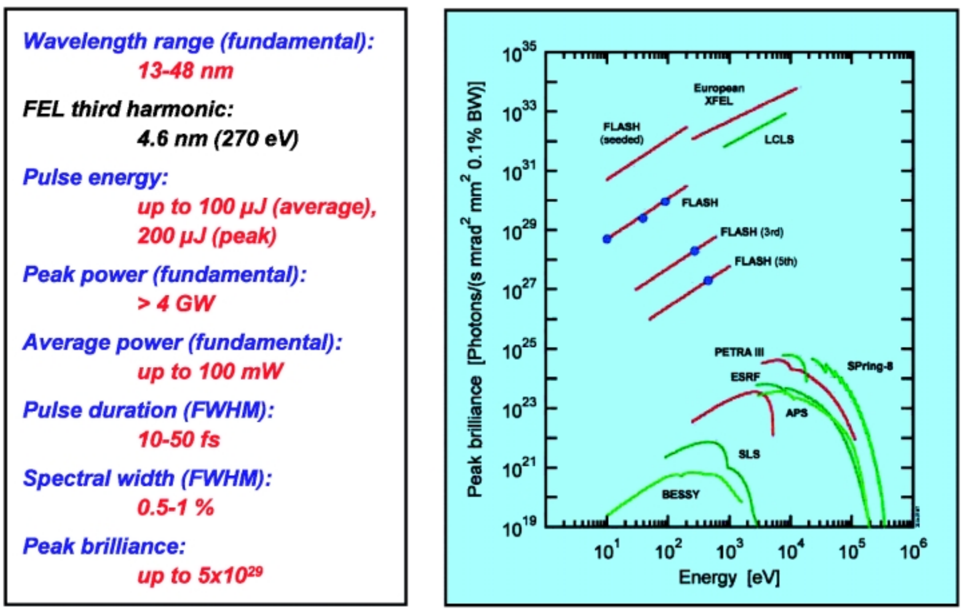

Fig. 6 Performance of the FLASH facility at DESY.

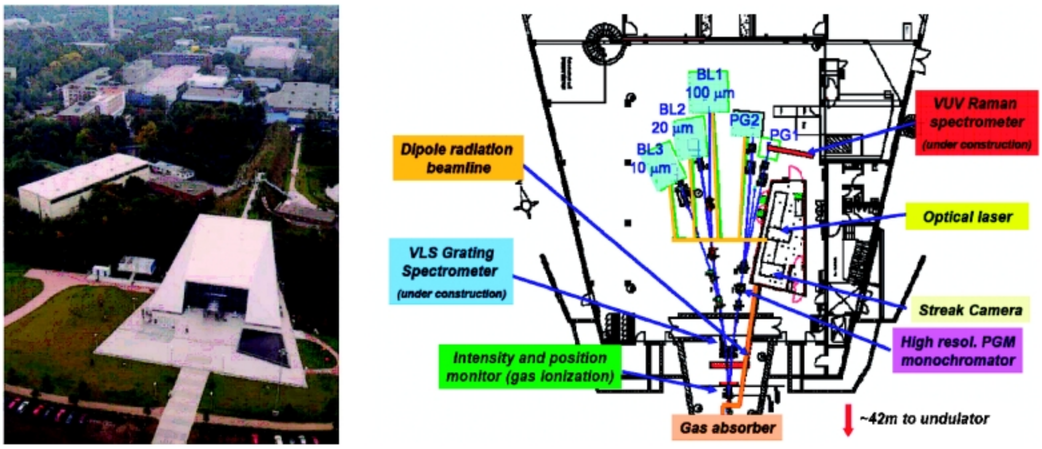

Fig. 7 Layout of the FLASH experimental hall.

into the water window–a wavelength range that is crucially important for the investigation of biological samples.

In the FLASH experimental hall the FEL beam is distributed to 4 experimental stations which are used one at the time (**Fig. 7**). In general the beamtime is shared in 12 hours shifts between two experiments performed at different stations. Three beamlines get the direct beam without downstream monochromator and focused to different spot sizes, the smallest focus reached so far was $\sim 2\,\mu$m in diameter. The 4^{th} beamline has a monochromator which will also serve a 5^{th} station where a Raman spectrometer is currently under construction. Photon diagnostics allow in a shot by shot mode the measurement of the beam intensity, the spectral distribution and the degree of coherence. An optical laser for pump & probe experiments is housed in a special hutch in the experimental hall together with a streak camera for determination of the jitter between FLASH pulses and the optical laser. The dipole radiation produced in the magnet tilting the electron beam into the dump is transported into the laser hutch and serves as reference signal.

Recently DESY published a brochure on FLASH describing the novel technologies and the free-electron laser principle on a level which should be useful to graduate students if they want to understand things beyond pure excitement, but also to scientists who may become interested in using this new tool for their own research. It is available on the internet:
http://hasylab.desy.de/sites/site_hasylab/content/e76/e6757/e6799/column-objekt17597/lbox/infoboxContent17599/FLASH-BroschrefrsWeb_2007-05-22_eng.pdf

4. Examples of science at FLASH

FLASH is not only a test bed for free-electron laser technology but also offers the users the opportunity to get familiar with the enormous scientific potential of

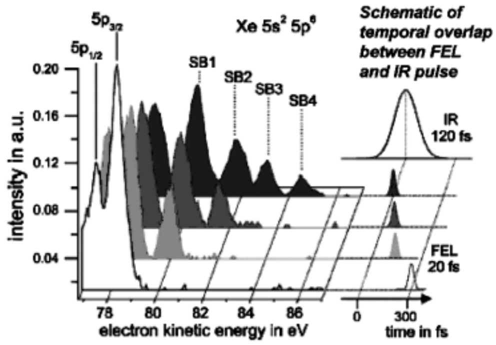

Fig. 8 Single-shot spectra in the region of the Xe $5p$-1 line and the high-energy sidebands taken at the nominal temporal overlap $\Delta t = 0$ and due to jitter also at $\Delta t \neq 0$. The spectra are normalized to total FEL + IR intensity.

these facilities and to learn how to perform experiments, which is fundamentally different from what is done at synchrotron radiation sources today. FLASH offers about 3000 hours dedicated user time per year, the facility is currently overbooked by a factor of 3.

Single-shot characterisation of independent femtosecond extreme ultraviolett free-electron and infrared laser pulses P. Radcliffe et al., Appl. Phys. Lett. **90** (2007) 131108

Many physical, chemical and biochemical processes happen so fast that it is only possible to uncover the intermediate steps using femtosecond light sources. Pump and probe experiments using X-ray FELs may uncover the dynamics of ultra fast processes at the atomic level. The key to success is the ability to synchronize the subpicosecond pump and probe pulses.

Two-color above threshold ionization of helium and xenon has been used to analyze the synchronization between individual pulses of the femtosecond extreme ultraviolet XUV free-electron laser FLASH and an independent intense 120 femtosecond mode-locked Ti:sapphire laser. Characteristic sidebands appear in

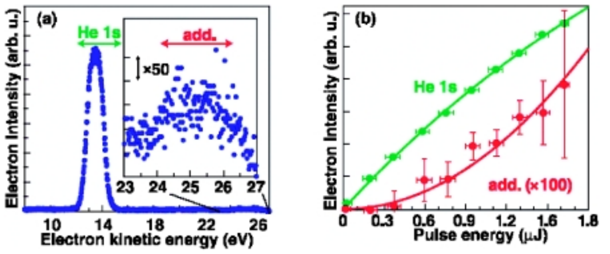

Fig. 9 Fluence dependent photoemission of helium with FEL radiation at $h = 38.5 \pm 0.2$ eV photon energy. (a) Photoelectron spectrum collected at photon pulse energies between 0.65–1.63 J/pulse in a 200 μm^2 focal spot. Inset: additional spectral features between 24–26 eV kinetic energy, not accounted for in single-photoemission. (b) Fluence dependence of the He 1s photoemission upper green circles and spectral features induced by resonant two-photon absorption lower red circles.

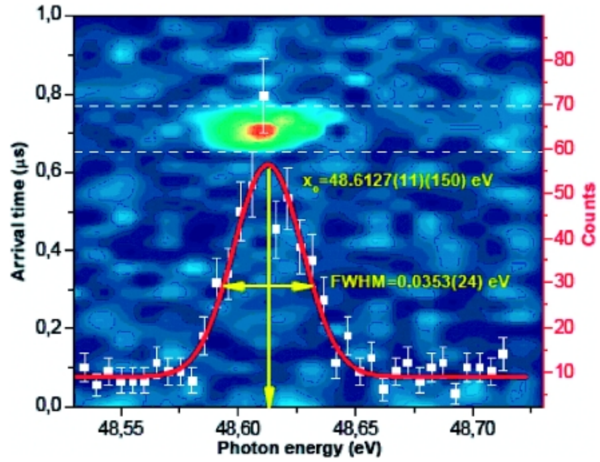

Fig. 10 Fluorescence spectra as a function of arrival time relative to the FEL pulse and versus photon energy.

the photoelectron spectra when the two pulses overlap spatially and temporally (**Fig. 8**). Up to four sidebands have been observed for the excitation of Xe at 13.8 nm. The cross-correlation curve points to a 250 femtosecond rms jitter between the two sources at the experiment. Numerical simulations combined with single-shot sideband intensity data yield a precision of better than 50 femtoseconds in the time window where both pulses are temporally overlapped.

Resonant two-photon absorption of extreme ultraviolet FEL radiation in Helium M. Nagasono et al., Phys. Rev. A **75**, 051406(R) (2007)

Understanding the interaction of the high intensity, short pulse radiation from free-electron lasers with matter is of crucial importance for whatever will be done with these new X-ray sources. In a photoemission experiment the nonlinear response of Helium to intense extreme ultraviolet radiation from FLASH was investigated (**Fig. 9**). Above the 1s line at electron kinetic energies around 13.5 eV one observes a spectral feature between 24 and 26 eV which shows a quadratic fluence dependence. The feature is explained as a result of subsequent processes involving a resonant two-photon absorption process into doubly excited levels of even-parity (N = 5 and 6), radiative decay to the doubly excited states in the vicinity of the He+ (N = 2) ionization threshold and finally the photoionization of the inner electron by the radiation of the next microbunches. This observation suggests that even parity states, which have not been measured using synchrotron radiation due to low pulse energy, can be investigated with intense radiation of FLASH. This also demonstrates a first step to bring nonlinear spectroscopy into the XUV and soft-X-ray regime.

Soft X-ray laser spectroscopy on trapped highly charged ions at FLASH S. W. Epp et al., Phys. Rev. Lett. **98**, 183001 (2007)

Highly charged ions are abundant in the universe, but virtual no experimental data exists on their interactions with energetic photons. FLASH and future FELs may determine the electronic structure of highly charged ions with unprecedented precision providing critical input to stellar models and enabling high accuracy tests of the theory of quantum electrodynamics, QED.

Fe^{23+} ions with a density of 10^{10} ions/cm^3 were produced in a new electron beam ion trap (EBIT) installed at FLASH. Fe^{23+} has only three electrons left and resembles lithium apart from the much stronger electromagnetic field experienced by the electrons. As a first objective the transition between the $1s^2 2s\ ^2S_{1/2}$ ground state and the exited $1s^2 2p\ ^2P_{1/2}$ state occurring in all three-electron ions from lithium to U^{89+} were investigated. The experiment was performed in a single photon resonant excitation scheme by tuning monochromatic FEL pulses through the resonance of this transition in Fe^{23+} at an energy of 48.6 eV corresponding to a wavelength of 25.5 nm. The fluorescence photon yield emitted upon relaxation of the excited state was registered as a function of the laser wavelength (**Fig. 10**).

According to QED theory various processes contribute to the total transition energy: Inter-electronic interactions, single electron one loop terms comprising self energy and vacuum polarization and the corresponding screening terms. The relative accuracy achieved in the Fe^{23+} experiment is already higher than the theoretical uncertainties, and the present experimental accuracy at FLASH allowed verification of the leading two-photon QED terms. Any further increase in the experimental precision will provide a systematic sensitivity improvement that may enable verification of the other processes contributing to the total transition energy.

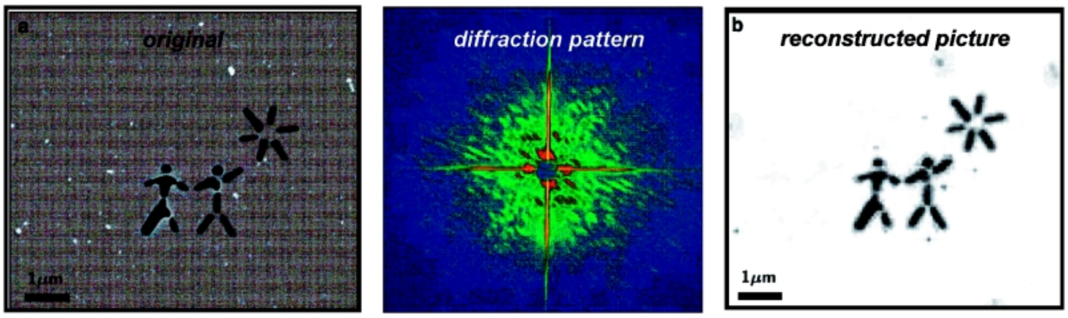

Fig. 11 Single shot diffraction pattern taken with 32 nm FEL radiation from FLASH and reconstruction.

Femtosecond diffractive imaging with a soft-X-ray free-electron laser H. N. Chapman et al., NATURE Physics **2**, 839 (2006)

Theory predicts that, with an ultrashort and extremely bright coherent X-ray pulse, a single diffraction pattern may be recorded from a large macromolecule, a virus or a cell before the sample explodes and turns into a plasma. The first experimental demonstration of this principle using the FLASH soft-X-ray free-electron laser has been achieved by an international collaboration lead by H. N. Chapman and J. Hajdu. An intense 25 femtosecond, 4×10^{13} Wcm^{-2} pulse, containing 10^{12} photons at 32 nm wavelength, produced a coherent diffraction pattern from a nanostructured non-periodic object, before destroying it at 60,000 K. A novel X-ray camera assured single-photon detection sensitivity by filtering out parasitic scattering and plasma radiation. The reconstructed image, obtained directly from the coherent pattern by phase retrieval through oversampling, shows no measurable damage, and is reconstructed at the diffraction-limited resolution (**Fig. 11**). A three-dimensional data set may be assembled from such images when copies of a reproducible sample are exposed to the beam one by one.

The experiment is considered a very important step on the way to single molecule imaging which is one of the most exciting possible applications of X-ray free-electron lasers. More recently a single shot ~10 femtosecond diffraction pattern of a living picoplankton organism could be recorded at a wavelength of 13.5 nm. The cell was injected into vacuum from solution, and shot through the beam at 200 m/s. Diffraction patterns are obtained only when the target particle and the photon flash overlap in space and time and therefore the criterion for success is the luminosity achieved in the experiment. Production of single particles and to focus and transport them to the interaction point is difficult so that the target density will be extremely low in most cases. To compensate for this one needs not only the extreme peak brilliance characteristic for all FELs but also a very high average brilliance, which can only be obtained using a linear accelerator in superconducting RF technology. This is one of the unique selling points of the XFELs as developed at DESY.

5. Concluding remarks

X-ray free-electron lasers will allow for the first time to study matter with atomic resolution in space and time which opens revolutionary new opportunities to investigate the dynamics of physical and biological systems. Instead of taking pictures we shall record movies, we want to see molecular machines at work. FELs do not work in an equilibrium mode as storage ring facilities do and each photon flash has to be characterized with respect to intensity, position, spectral distribution and degree of coherence. Therefore the development of shot by shot photon diagnostics is of key importance for successful research with FELs. In many cases samples will be different from what we know from synchrotron radiation research and a lot of effort has to be put in sample preparation, alignment and the development of special sample environments. For each shot all beam and sample parameters have to be stored in the computer together with the large amount of data from area detectors. The data rate will be enormous taking into account that e.g. at the European XFEL Facility 30,000 flashes per second are expected, each of them may produce a fully interpretable diffraction pattern. Because of the complexity of the experiment and the different expertise's needed, larger collaboration will be formed around the different types of experiments. The interpretation of the data will probably be done in a similar way as known from particle physics. The hypothesis for the interpretation of an experiment will be checked by doing cuts through a large multi parameter data set, most of that will be done in the home laboratory. Making use of modern data handling techniques like GRID the data will be accessible to all members of the collaboration and exchange of results among them will be easy.

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