Eur. Phys. J. D **36**, 193–197 (2005)
DOI: 10.1140/epjd/e2005-00268-4

THE EUROPEAN PHYSICAL JOURNAL D

Investigations of ultrafast phenomena in high-energy density physics using X-ray FEL radiation

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Received 31 May 2005

Abstract. The advent of highly intense and ultrashort pulses of short-wavelength radiation in the vacuum-ultraviolet to X-ray regime provides for the first time the possibility to study plasmas at the time scale of equilibration or even electron thermalization. The emerging radiation sources are free-electron lasers (FEL) based on high-energy electron accelerators. FELs provide a peak brilliance nine orders of magnitude higher than the best performing X-rays sources today. The FEL radiation parameters will enable the creation of high-energy density states of matter and the development of new diagnostics tools to investigate dense plasmas. As the first of the new sources the VUV-FEL at DESY, Hamburg becomes operational for high-energy density physics experiments during 2005.

PACS. 41.60.Cr Free-electron lasers – 42.55.Vc X-ray and γ-ray lasers – 52.40.Db Electromagnetic (nonlaser) radiations interactions with plasma – 52.50.-b Plasma production and heating – 52.70.La X-ray and γ-ray measurements

1 Introduction

Optical laser experiments in the field of plasma physics have received considerable attention due to the availability of high-intensity lasers. A particular breakthrough was achieved using lasers with a pulse duration of 100 fs and below [1]. These ultrashort pulses allow to disentangle the excitation of the plasma from its temporal evolution. Using pump-probe techniques it thus becomes possible to study plasma parameters time-resolved. Since matter with electron densities near or larger than the critical electron density $n_c = 1.11 \times 10^{21}$ cm$^{-3}/\lambda^2[\mu$m] becomes opaque to radiation of wavelength λ optical laser radiation is not transported in dense matter and experiments are usually restricted to gases. Intense radiation is absorbed at the surface and heat propagation and shock fronts can lead to plasma formation. The induced time- and space-dependent formation of transient states of matter makes the description of the sample system and the formation of plasma states quite demanding.

In contrast, X-ray radiation with much higher critical densities ($n_c = 10^{29}$ cm^{-3} for 12 keV radiation) penetrates homogeneously into dense matter. X-ray methods to study high-energy density physics therefore play an important role. However, X-ray radiation parameters of present day sources do not fit the requirements of plasma physics experiments to employ single pulses to either heat, scatter, or probe matter that is transient. The best performing X-ray sources today are synchrotron radiation in the soft to hard X-ray regime, high harmonic generation and X-ray lasers in the soft X-ray regime, and ultrashort pulse laser-based Kα X-ray sources. None of these sources provides simultaneously high pulse intensity, ultrashort pulse duration, and high brilliance required for application of X-ray methods in high-energy density physics described below. Accelerator-based free-electron lasers (FEL) will change this situation drastically by providing tunable, narrow-band, and ultrashort pulses of very high intensity in a wavelength range from the VUV to the hard X-ray regime. FEL sources currently become available for experiments in the soft X-ray regime (10–200 eV) and construction of facilities for the hard X-ray regime has started.

High-energy density physics includes the investigation of matter and radiation under extreme conditions and covers the regimes of warm and hot dense matter. X-ray FELs will provide the possibility to generate dense plasmas with temperatures >100 eV and pressures >10^4 GPa [2]. Moreover, FELs provide an excellent tool for the spectroscopic investigation of plasma states at or near solid density [3]. Diffraction of hard X-rays enables atomic resolution in determining interatomic distances and structure of the plasma [4]. X-ray FEL radiation applied in pump-probe experiments is thus a unique tool to gain time-resolved information on parameters of dense plasmas which are otherwise opaque to optical probing.

In the next section the development of X-ray FEL radiation sources and their current status will be described before introducing the advantages of using these sources in investigations of high-energy density physics. The last section discusses time-resolved experiments to investigate ultrafast phenomena in high-energy density physics.

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Table 1. Properties of X-ray SASE FEL radiation for a set of photon energies. Parameters for 8–12 and 0.8–3 keV correspond to the hard X-ray FEL projects Linac Coherent Light Source (LCLS) [15] and European XFEL [16]. The values for 0.2 and 0.04 keV have been calculated for the VUV-FEL in short-wavelength and femtosecond mode of operation, respectively. Power densities are calculated for a focal spot size of 10 μm neglecting reflectivity and acceptance losses.

Photon energy (range) Wavelength (range)	keV nm	8–12 [15,16] 0.1–0.15	0.8–3 [15,16] 0.4–1.5	0.2 [17] 6.4	0.04 [18] 30
Pulse duration (FWHM)	fs	100	100	200	50
Rel. bandwidth (FWHM)	%	0.1	0.3	0.36	0.8
Source size (FWHM)	μm	100	80	140	270
Divergence (FWHM)	μrad	1	9	24	80
Pulse intensity	phts	10^{12}	2×10^{13}	1.8×10^{13}	1.5×10^{13}
Pulse energy	mJ	2	4	0.6	0.1
Peak power density	W/cm^2	2×10^{16}	4×10^{16}	3×10^{15}	2×10^{15}

2 X-ray FEL source properties

Before discussing the properties and status of X-ray FEL sources a review of the limitations of current synchrotron radiation sources will highlight the advances obtained by the new sources. Synchrotron radiation is an outstanding tool to investigate static properties of matter, but the investigation of kinematic processes in the time domain is limited. Most time-resolved experiments using synchrotron radiation exploit the time domain above milliseconds and time resolution is obtained by utilizing adequately fast detectors. Time-resolved experiments in the picosecond to nanosecond time domain employ pump-probe studies using optical laser as the pump [5]. These experiments achieve time resolution by varying the delay between pump and probe pulses limited by the duration of the X-ray pulses of the order of 100 ps. Single pulse intensities from 10^5 to 10^{10} photons can be obtained at the sample position, depending on bandwidth and operating mode [6].

The X-ray pulse duration of synchrotron radiation is limited by the electron bunch length, a characteristic property of the storage ring resulting from the equilibration of energy losses due to radiation and acceleration by radio-frequency fields. The electron bunch length varies for third-generation synchrotron sources from 3 to 30 mm, leading to respective X-ray pulse durations of 10 to 100 ps [7]. A special operating mode of the storage ring allows to produce much shorter electron bunches providing few ps X-ray pulse durations [8]. This mode allows only small charge per bunch and the pulse intensity therefore is small. A more attractive method to generate ultrashort pulse in storage rings utilizes modulating the electron beam energy by interaction with a co-propagating ultrashort optical laser pulse [9]. X-ray pulse durations of few 100 fs and intensities of the order 10^7 photons/s can be expected from these ‘femtosecond slicing’ sources.

Ultrashort electron bunches with a charge of the order of 1 nC can only be produced in linear accelerators using special bunch compression schemes, like applied for the Sub-Picosecond Pulse Source (SPPS) at SLAC, Stanford [10]. Electron bunches with a length of <30 μm are utilized in an undulator to produce X-ray synchrotron radiation pulses with 80 fs duration [11]. Pulse intensities of 10^7 photons have been successfully used in laser/X-ray cross-correlation experiments [12]. Linear accelerator-based FELs use this spontaneously emitted synchrotron radiation to generate a radiation field with which the co-propagating electron bunch interacts. Providing a very low emittance electron beam this interaction leads to coherent emission of radiation with pulse intensities of 10^{12}–10^{14} photons and pulse durations of 30–200 fs. Several FEL schemes have been proposed to generate X-ray radiation, but the most mature method today is the self-amplified spontaneous emission (SASE) scheme [13]. This scheme is applied at the VUV-FEL at DESY, Hamburg, the first soft X-ray FEL becoming operational [14], and will be used for the hard X-ray FELs in Stanford and Hamburg. Table 1 indicates typical parameters for SASE FEL radiation at different X-ray photon energy ranges.

The FEL photon energy coincides with the fundamental harmonic of the undulator. The total radiation spectrum is a superposition of FEL radiation and of spontaneously emitted synchrotron radiation, the latter shown in Figure 1. Taking electron beam parameters required for SASE the spontaneously emitted synchrotron radiation (SR) is collimated in angle and bandwidth nearly as good as the FEL radiation. Figure 1 shows that SR is emitted only at photon energies corresponding to the fundamental and higher harmonic lines. Relative intensities of SR in the fundamental and higher harmonic lines are indicated in Table 2. In addition, for specific settings of the SASE undulator 2nd and 3rd harmonics of FEL radiation will be observable at typically 10^{-3} of the fundamental FEL line [20].

The VUV-FEL provides SASE FEL radiation for photon energies from 20 to 200 eV in the fundamental FEL line by tuning the electron energy from 0.33 to 1 GeV, respectively. Using 2nd and 3rd harmonics this range can be extended to 600 eV. The super-conducting electron accelerator enables to generate trains of up to 7200 electron bunches with 10 Hz repetition rate. This pattern will enable experiments requiring very high average intensities integrating many pulses, but also fast feedback methods for electron beam stabilization will thus become feasible. The FEL undulator consists of six 4.5 m long modules

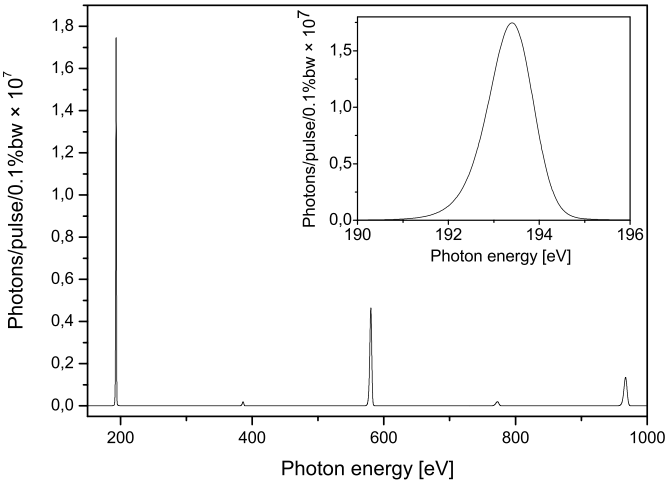

Fig. 1. Spectral distribution of spontaneously emitted synchrotron radiation spectra for the VUV-FEL parameters at 193 eV FEL radiation. Numbers correspond to angular integrated flux assuming full acceptance. The insert shows the profile of the fundamental line.

Table 2. Relative intensity of spontaneously emitted synchrotron radiation normalized to the fundamental FEL line. Calculations using the SPECTRA code [19] did not take broadening due to electron energy losses and diffraction effects in the long undulators into account. The experimentally observable ratios will therefore decrease with respect to tabulated values.

FEL line	1st harm.	3rd harm.	5th harm.
200 eV (VUV-FEL)	10^{-6}	3×10^{-7}	10^{-7}
12.4 keV (XFEL)	3.9×10^{-4}	2.8×10^{-4}	2.1×10^{-4}

providing about 1000 undulator periods of 27.3 mm length. For a gap of 12 mm a peak magnetic field of 0.47 T is achieved. The VUV-FEL is currently commissioned and first experiments have started in summer 2005. FEL radiation will be provided by a beam transport system to presently five beamline ports, two using a high-resolution plane grating monochromator and three using purely mirror reflections. Focussing optics produce focal spots from 10 to 100 μm at the various end-stations. The experimental set-ups including vacuum chamber, sample preparation and detection systems will be provided by the user groups. The facility includes diagnostics for pulse energy, beam position and beam pointing on a shot-by-shot basis. The spectral distribution is at present measured by a spectrometer intercepting the FEL beam. The measurement of timing features, like pulse arrival time and temporal distribution, remains a new field for ultrashort X-ray radiation (see e.g. [11]) and will be a key development during the startup of operation of the VUV-FEL. The facility includes an optical laser system to be used for FEL diagnostics measurements, using cross-correlation techniques, and for pump-probe experiments. The optical laser can provide the same pulse pattern as the accelerator.

3 High-energy density physics applications

The high pulse intensity, ultrashort pulse duration, high photon energy, and extreme brilliance of X-ray FEL radiation provide for high-energy density physics unique possibilities to study dense plasmas. Here, we present two areas of application and the following section discusses aspects of time-resolution in these experiments.

The first area of new experiments arises from the possibility to use FEL radiation to generate high-energy density states of matter. X-ray radiation is absorbed in a wide range by photoionization raising electrons directly to continuum levels. Depending on the energy difference $\Delta E = E_x - I_o$ for the ionization potential I_o the initial electron energy is defined. In dense matter the photoelectrons equilibrate almost instantaneously on timescales of few fs. The relaxation between electrons and ions takes several 100 fs and hydrodynamic expansion sets in after 1 ps [2]. During heating the density therefore remains constant. This isochoric heating corresponds in the density-temperature and density-pressure phase diagrams to a vertical line, meaning a jump in temperature respectively in pressure at constant density. Temperatures up to several 100 eV and pressures up to 10^5 GPa have been calculated [2]. Exponential absorption of X-rays leads to slabs of uniformly heated matter. The heating process depends on ΔE and therefore on the exact choice of material and photon energy [21]. Adjustment of the deposited energy is possible by adjusting the radiation energy and photon energy, and through the choice of sample material and defines the initial plasma temperature and pressure.

From the initially generated plasma state the system expands following a thermodynamically determined path in the phase diagram. Varying initial parameters the relaxation paths covers the entire temperature-density phase diagram from ~100 to below 1 eV and densities decreasing from solid density. Measurements of the opacity and other equation of state properties during this relaxation will provide new, and not otherwise obtainable data. The expansion may also lead into the instable liquid-gas two-phase regime in the phase diagram. Experimental data for the liquid-gas two-phase regime including critical point data are extremely scarce. Furthermore, solid density plasmas at temperatures $\leq$10 eV can be generated in a well-defined way. This regime, also called warm dense matter, describes the transition from condensed matter to ideal plasmas and is yet very difficult to produce. Whereas the condensed matter regime is dominated by short- and long-range order effects, ideal plasmas are well described by isolated ions inside a thermal bath. In the warm dense matter regime both descriptions fail and correlations have to be integrated into plasma theories. Experimental data obtained by X-ray FELs will be crucial for bench-marking of new theories for the warm dense matter regime [3] and for the investigation and understanding of high-energy density matter in general. A particular field of application are astrophysics problems, e.g. planet interiors or formation.

In the second area of experiments the tunable, small bandwidth, and intense FEL radiation is used for spectroscopic experiments to diagnose dense plasmas. The methods of X-ray Thomson scattering and FEL-induced emission spectroscopy seem particular promising here. X-ray Thomson scattering enables the measurement of the plasma temperature, density, charge state, and collective properties such as ion-acoustic or electron plasma waves in dense plasmas [22,23]. The technique requires a spectrally resolved measurement of the scattered photons at a fixed scattering angle θ between incident and emitted photon momenta. For scattering from both free and weakly bound electrons the final state is a free electron and spectra are shifted in energy $E_x' = E_x/(1+E_x(1-\cos\theta)/mc^2)$ with the electron rest mass mc^2. The measured spectra are analyzed to distinguish scattering from free, weakly bound, and tightly bound electrons. The spectral distribution of the free electron component reveals details of the electron velocity distribution, and therefore of the temperature distribution. The result depends in general on the dynamical structure factor $S(q,E_x)$, with the momentum transfer q, and therefore on details of the plasma interaction [24]. Coherent scattering allows to investigate collective plasma modes [25]. An estimation of the scattering conditions for X-ray Thomson scattering using the VUV-FEL can be found in reference [26].

FEL-induced emission spectroscopy requires tunable, intense, and small bandwidth radiation to pump line transitions in dense plasmas [3]. The enhancement of excited state populations will modify the plasma emission that is detected spectrally resolved. Using this method kinetic rates or populations can be investigated. A particular application is the investigation of inversion mechanisms for the production of X-ray laser schemes. Here, X-ray FEL radiation can produce extreme non-equilibrium conditions during and immediately after irradiation. Using the VUV-FEL photoionization of He with ΔE greater but near I_o leads to a small electron temperature, and therefore to a high recombination rate to excited atomic states. Simulations for 25 eV photon energy interacting with He gas at 10^{19} cm^{-3} show inversion of the He ^{1}P$_1$ state by three-body recombination and lasing at the He-α line (21 eV) with a gain up 100 cm^{-1} [27]. He-α line lasing has not been observed yet since no efficient pumps were available. The proposed studies will enable for the first time to study experimentally important atomic processes, level kinetics, line shape formation, or X-ray laser gains. An evaluation of radiative pumping rates for X-ray FEL experiments can be found in reference [3].

4 Investigation of ultrafast phenomena

In the following we discuss time-resolved experiments to investigate plasmas, in particular in the context of the VUV-FEL. The availability of FEL pulses of duration of 100 fs and below and of optical laser pulses of similar duration will enable pump-probe techniques reaching temporal resolutions of a few 100 fs. To achieve such time-resolution either synchronization of the two light sources of 100 fs and better is needed or recently tested measurement techniques of the delay between pump and probe pulses [11] must be established. Pump-probe experiments using FEL and optical laser pulses could be applied both ways. Creating dense plasmas using the optical lasers leads to inhomogeneous and highly dynamical processes difficult to analyze using optical wavelengths. Probing using spectroscopic techniques based on X-ray FEL radiation (compare Sect. 3) will therefore provide unique possibilities for the analysis of these plasmas states. Delaying the probe pulse enables investigation of plasma parameters at varying times during evolution and/or relaxation of the plasma. Vice versa, the full potential of well-established optical laser techniques can be exploited in FEL radiation pump / optical laser probe experiments. An example is to study surface expansion using frequency domain interferometry [28].

A different approach for time-resolved experiments employs pump and probe pulses both provided by the FEL. Typical ratios for the intensity of spontaneous radiation and higher harmonic lines with respect to the fundamental FEL lines have been indicated in Section 2. SR and higher harmonic FEL radiation are reduced in intensity by several orders of magnitude with respect to the fundamental FEL radiation. In pump-probe experiments they are likely to be used as probe pulse. Special X-ray optics are required for splitting and delaying the two beams (see e.g. [29]). Depending on the requirements of the experiment filters or gratings can be employed to separate the two photon energies [30]. Originating from the same electron bunch the two pulse will be naturally synchronized. An application of this technique will be the investigation of plasma evolution using the fundamental FEL radiation to heat solid matter to temperatures of a few eV and probe the plasma by Thomson scattering employing the 3rd harmonic line. Using FEL radiation with a photon energy of 200 eV and pulse energies of 100 μJ and applying a special beam geometry with moderate focusing to 30–100 μm will lead for Al samples to temperatures of 10 and 1 eV, respectively [26]. Third harmonic FEL radiation with a photon energy of 600 eV will provide an excellent probe beam for Thomson scattering to investigate plasma ionization, temperature, and collective phenomena. Special high-throughput optics will be required for separation of the fundamental and 3rd harmonic radiation and for focussing both radiations on the sample. Delays will be achieved by variation of the optical path difference.

The third possibility to investigate time-dependent phenomena in high-density energy matter is to use ultrafast X-ray streak cameras. These cameras reach sub-picosecond time resolution [31] and will be used in combination with spectrometers to monitor spectrally resolved self-emission from the plasma [32]. The time-resolved data will enable the investigation of temperatures and plasma kinetics at ps time-scale.

5 Summary

Utilizing radiation provided by X-ray FEL sources with extremely high peak brilliance new possibilities for high-energy density research will be provided in at least two important areas. Firstly, the ultrashort duration X-ray radiation enables to generate plasma states of matter at or near solid density and temperatures from ∼1 to 100 eV by isochoric heating. Secondly, the high intensity in combination with the high brilliance enables to apply new spectroscopic techniques for plasma diagnostics. The intrinsic time resolution of the ultrashort pulses and the application of pump-probe schemes will enable the investigation of ultrafast phenomena. First experiments have started during 2005 using radiation at 40 eV. These experiments are extended to photon energies of 200–600 eV during the following years before the first hard X-ray experiments will become possible by 2009.

The authors are grateful to H. Schulte-Schrepping for calculating the contribution of spontaneous radiation by the FEL undulators.

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