

Soft x-ray scattering using FEL radiation for probing near-solid density plasmas at few electron volt temperatures

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Abstract

We report on soft x-ray scattering experiments on cryogenic hydrogen and simple metal samples. As a source of intense, ultrashort soft x-ray pulses we have used free-electron laser radiation at 92 eV photon energy from FLASH at DESY, Hamburg. X-ray pulses with energies up to 150 μ J and durations 15-50 fs provide interaction with the sample leading simultaneously to plasma formation and scattering. Experiments exploiting both

of these interactions have been carried out, using the same experimental setup. Firstly, recording of soft x-ray inelastic scattering from near-solid density hydrogen plasmas at few electron volt temperatures confirms the feasibility of this diagnostics technique. Secondly, the soft x-ray excitation of few electron volt solid-density plasmas in bulk metal samples could be studied by recording soft x-ray line and continuum emission integrated over emission times from fs to ns.

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1. Introduction

Short-wavelength radiation exhibits significant differences in its interaction with matter compared to near-visible radiation. This had led to the proposal to employ intense, ultrafast, tunable and bright x-ray sources in studying properties of dense plasmas at an un-paralleled level of detail [1]. Several studies suggested the creation of solid density plasmas by intense femtosecond x-ray pulses [2, 3] or the probing of dense plasmas using various x-ray scattering and spectroscopy techniques [4-7]. Since existing conventional sources could not provide x-ray radiation with the required properties these experiments were, until recently, not possible. The first facility to provide intense, femtosecond, high-brightness, soft x-ray radiation is the free-electron laser (FEL) facility FLASH at DESY, Hamburg starting user operation in 2005 [8, 9]. In this paper we report on two types of soft x-ray plasma physics experiments, both carried out for the first time and both using 92 eV FEL radiation. These experiments aim at the investigation of thermodynamic properties such as electron temperature, electron density and ionization for strongly coupled plasma systems. In particular, the regime of interest is near solid electron densities of $n_e = 10^{21} - 10^{23} \text{ cm}^{-3}$ and temperatures from a few to tens of electron volts.

In contrast to ideal plasmas, for strongly-coupled plasmas correlations between the charged particles have to be considered. The regime of strongly-coupled plasmas can be identified by the ratio of Coulomb energy to the thermal energy, i.e., the dimensionless coupling parameter $\Gamma = e^2 / (4\pi\epsilon_0 \bar{d} k_B T_e)$, being larger than unity, where $\bar{d} = (4/3 \pi n_e)^{-1/3}$. The interest in studying these systems experimentally arises from the complexity of the theoretical description and the necessity to verify the applicability of models using experimental data. Experiments have been limited so far by the fact that near-visible laser

methods bear intrinsic restrictions for the investigation of near-solid density matter. When using near-visible radiation the probing of dense matter is not possible due to strong absorption and reflection. Further, the generation of plasmas using high intensities is accompanied by complex shock wave creation as well as strong density and temperature gradients. X-rays overcome both problems due to their largely increased penetration power.

2. Description of experimental techniques

2.1. Measuring near-solid density plasma parameters using inelastic soft x-ray scattering

The measurement of plasma parameters such as temperature, free electron density or ionization is required for the investigation of equation-of-state properties. One standard technique is Thomson scattering using near-visible radiation [10]. The applicability of this method is limited to plasmas with much smaller densities than targeted here. For near-solid density plasmas x-ray radiation is required and the technique of x-ray Thomson scattering had been successfully introduced using few keV x-rays, generated by high-energy laser radiation interaction with backlighter samples [11, 12]. Since FEL pulses provide similar photon numbers per pulse as used in the reported high-energy laser experiments, but have an even higher brightness, it is conceptually straightforward to implement this inelastic x-ray scattering technique using these new sources. Moreover, the much higher repetition rates of the FEL, when compared to the high-energy lasers, further facilitate the investigation of various samples, sample configurations, as well as scanning wide parameter ranges. At the same time the high temporal resolution in FEL experiments enables one to investigate the dynamics of plasma formation and

equilibration. We report here initial experiments carried out in the soft x-ray regime at 92 eV photon energy.

The scattering kinematics in inelastic x-ray scattering is determined by the momenta of incoming x-ray photon \vec{k}_i and scattered photon \vec{k}_f , where the momentum $\vec{K} = \vec{k}_i - \vec{k}_f$, $|\vec{K}| = K$ is transferred to the scattering particle. The kinematics determines if the collective plasmon-like character of electron excitations or non-collective single electron properties are probed. To distinguish the two regimes the scattering parameter $\alpha = 1/(K\lambda_{D,sc})$ is used. $\alpha > 1$ corresponds to the collective regime where the probed length scales are larger than the Debye screening length $\lambda_{D,sc}^2 = \epsilon_0 k_B T / (n_e e^2)$. Here, scattering from collective electron excitations, i.e., plasmons, can be observed. In the non-collective regime, $\alpha < 1$, the probed length scale is shorter than $\lambda_{D,sc}$ and the momentum distribution for individual electrons is probed. The energy transfer from the photon to the single electron is $\hbar\omega = \hbar(\omega_i - \omega_f)$. For bound electrons with $\hbar\omega \geq E_B$, E_B being the binding energy of this electron, Compton scattering becomes observable. Since the energy transfer in the soft x-ray regime is very small, typically below the energy resolution and difficult to investigate, these first inelastic x-ray scattering experiments using FEL sources have been carried out in the collective scattering mode. In this mode scattering from the plasmon gives rise to down- and up-shifted resonances, relative to the elastic scattering. From the energy difference of the resonances with respect to the FEL probe pulse the electron density can be estimated and the slope of the peak heights of the resonances allows an independent estimate of the electron temperature [7]. Experimentally, an infrared femtosecond laser is used to heat a liquid hydrogen sample

with an initial density below solid-density. We estimate that the plasma equilibrates within a few picoseconds, before the measurement of plasma parameters by soft x-ray scattering occurs. Using two soft x-ray spectrometers at different scattering angles θ , the simultaneous measurement of scattering for two values of α , respectively K , provides the possibility of investigating the scattering from electrons bound to the ions [13].

2.2. Creation of solid-density plasmas using soft x-ray radiation

The current operation of FLASH allows one to obtain intensities up to 10^{17} – 10^{18} Wcm⁻² by focusing the soft x-ray FEL beam. The interaction of the focused soft x-ray radiation with a small volume of solid-density matter can lead to an energy deposition of up to $\sim 10^3$ kJ/cm³ and subsequent plasma formation. The energy absorption occurs over the duration of the FEL pulse of few ten femtoseconds, which is too short for the material to expand. For our experimental conditions and sample materials the primary absorption process is photoabsorption by L-shell electrons. Initially, the material enters into a highly excited, non-equilibrium state that develops with time after the FEL impact. During the entire cycle the sample transits from strongly-correlated cold matter, via a highly-excited non-equilibrium plasma, to an equilibrated plasma state that cools down while expanding. Both continuum and line emission occur at different times in this cycle. The spectral shape of the continuum radiation and the relative intensities of plasma emission lines can be used to determine plasma parameters [14]. In these experiments one has to consider averaging effects due to spatially varying energy deposition and to time integration of the developing plasma states. Initially no time resolution could be applied to the spectral measurements. Therefore it is not possible to determine when radiation from the various emission channels occurred.

3. Experimental setup

Experiments have been carried out using soft x-ray radiation provided by FLASH, an FEL facility operating in the self-amplified spontaneous emission (SASE) mode [15]. In FLASH very low emittance electron bunches are generated in a photo-injector and are immediately accelerated to avoid space charge effects. After two compression steps using magnetic chicanes the electrons are accelerated to their final energy of 300 to 1000 MeV, producing soft x-ray FEL radiation, in a fixed gap 30 m long permanent magnet undulator, in the range 30 to 200 eV [16]. In these experiments we used 92 eV photon energy, which corresponds to an electron energy of 700 MeV. In the employed operation mode of FLASH the electron bunch, which has a higher density on the its leading edge, generates x-ray pulse durations of 15 – 50 fs with a bandwidth near 1.0 % and pulse energies up to 150 μ J. These x-ray pulses are transported to the experiments using carbon-coated mirrors operated at a grazing angle of 2 and 3 deg. The experiments have been carried out at BL2 of FLASH with two flat and one ellipsoidal mirrors, the latter producing a focal spot of 20 – 30 μ m diameter (FWHM). The total beamline transmission is considered to be $64\pm 4\%$ [16].

For the experiments reported here fluctuations of the pulse energy and of the spectral distribution need to be considered. The pulse energy I_o is measured pulse-by-pulse and this measurement is used for normalization of single-pulse data [17]. For measurements that integrate over several x-ray pulses the averaged pulse energy is used for normalization. The spectral distribution also fluctuates from pulse-to-pulse as is shown in Fig. 1. In the case shown, which has been collected during a particularly unstable period

of FEL operation, the averaged distribution exhibits a bandwidth of $\sim 2\%$ corresponding to 2 eV, and the individual spectra indicate a spectral structure with a typical peak width of 0.3–0.5 eV. Since only few modes are populated the weight of the spectral distribution can vary strongly from pulse-to-pulse. In order to normalize the scattering spectra with respect to this fluctuation an additional spectrometer measures the spectral distribution of the incident beam. Fluctuations of the temporal distribution of the FEL pulses can occur within an envelope of smaller 50 fs, but do not affect the data analysis presented here. FLASH can accelerate trains of 1-800 electron bunches with a repetition rate of 5 Hz. The repetition rate of bunches within the train can be selected between 40 kHz and 1 MHz. Most of the experiments reported here have used single pulses or a 5 Hz repetition rate. In a few cases trains with up to 30 pulses at 5 Hz, corresponding to 150 pulses per second have been used to increase the counting statistics.

The experimental setup is mounted in ultrahigh vacuum conditions using a total of 2200 l/s pumping speed. The geometry is chosen such that the soft x-ray spectrometers are mounted in the vertical plane at scattering angles θ equal to 0, 16 and 90 degree. The choice of the vertical plane follows from the very high degree of linear polarization of incident radiation in the horizontal plane. The spectrometer at $\theta = 0$ degrees is used for measuring the spectral distribution of the incident radiation. In flat-field geometry a planar variable line spacing grating with 1200 lines/mm is mounted at ~ 170 cm from the interaction point. A 100 μm slit is used for background reduction and vacuum separation. For detection a back-illuminated CCD is used and we obtain an energy resolution $\Delta E/E \cong 3.6 \times 10^{-3}$ [18]. The spectrometer at $\theta = 16$ degrees employs the same spectrometer principle and the same grating, which is mounted 23.5 cm from the interaction point. The

main spectrometer, mounted at $\theta = 90$ degrees, was optimized to provide large solid angle, high throughput and high spectral resolution in order to maximize the signal level. For the first experiment a focusing spectrometer based on a transmission grating was used [19], but artifacts arising from the support grid of the grating and the need for improved alignment and higher efficiency led to design and construction of a new spectrometer [20]. This spectrometer employs a toroidal mirror together with an 800 lines/mm variable line spacing reflection grating, accepts a solid angle of $19 \times 10^{-4} \text{ sr}$, obtains a resolution of $\Delta E/E \cong 5 \times 10^{-3}$ at 92 eV and has an estimated throughput of $\approx 4 \times 10^{-2}$ including detection efficiency of the back-illuminated CCD. In addition to the soft x-ray spectrometers we use optical spectrometers for detection of emission radiation in the near-visible regime. Analysis of line and continuum emission in this regime should provide further information about the temperature of the hydrogen plasma.

The samples are positioned in the focus of the FEL beam. The inelastic scattering experiment uses a liquid hydrogen jet produced by nozzles of 10 – 20 μm diameter at a temperature ~ 20 K [21]. The jet diameter under ideal conditions is equal to the nozzle of the liquid hydrogen source. The preparation of the liquid jet can lead to a variety of jet conditions. The phase space of liquid hydrogen is rather dense in this regime and small fluctuations of parameters like pressure or temperature and impurities will lead to rather strong variations. In particular, we have observed that the mean density of the liquid jet can vary strongly. In the ideal condition we observe a homogeneous jet of liquid hydrogen with a mass density $\rho = 0.0706 \text{ g cm}^{-3}$ [22]. In another operation mode the hydrogen is ejected in tiny droplets with liquid density forming a jet with a mean density approximately 5 – 10 times less than for the liquid. The solid samples are mounted on a

manipulator enabling translation to the chamber center once the liquid hydrogen source is retracted. The incidence angle of FEL radiation is roughly 45 degrees with respect to the sample surface.

The near-solid density hydrogen plasma is created using a femtosecond infrared laser delivering 3 – 5 mJ pulses at 800 nm at 5 Hz into a focal spot of ~ 30 μm diameter. A nearly collinear excitation geometry using a parabolic mirror with a central hole is chosen. With a measured pulse duration of roughly 150 fs obtained by compression of the stretched pulse just before entering the vacuum chamber this system reaches intensities of $\geq 10^{15}$ Wcm^{-2} [23]. The laser is synchronized using a radiofrequency clock to the arrival of the x-ray pulse with an accuracy of few ps over of several hours duration [24]. Sub-picosecond time resolution can be obtained in a pulse-by-pulse measurement of the temporal fluctuation of the infrared laser with respect to the x-ray arrival [25]. In the experiments reported here we use a time resolution of the order 1 – 2 ps obtained by cross-correlating the infrared laser to visible synchrotron radiation using a streak camera [24]. To improve the signal-to-noise ratio we always analyzed differences of two measurements: One with the infrared laser before the FEL, leading to plasma formation, and a second, with the infrared laser after the FEL, thus probing the cold hydrogen system. In addition, background images without x-ray illumination and of similar duration are taken to account for stray scattering and detector noise.

In the reported experiments x-ray scattering emerges from an interaction volume formed by the crossing of the soft x-ray beam of 25 μm (FWHM) diameter and the 20 μm diameter cryogenic liquid hydrogen jet. The incident intensity in this scenario is

$2.5 \times 10^{14} \text{ W cm}^{-2}$ corresponding to 30 μJ pulse energy at the sample, 25 fs duration and 25 μm focal spot diameter. The absorption length of 92 eV radiation in liquid hydrogen of 9.5 μm [26] corresponds to 12 % transmission for a 20 μm thick sample. For the measurements on solid aluminum and magnesium the FEL beam hits the surfaces at an angle close to 45 degrees and the absorption lengths are 0.036 μm and 0.039 μm , respectively. These absorption lengths are valid for the unperturbed ground-state materials. If the deposited energy is sufficient to modify the material properties significantly, one has to consider a transiently changing absorption. We have simulated this effect using the HELIOS code [27] assuming that the energy deposited by the FEL beam is absorbed through photo-absorption and inverse Bremsstrahlung. For the particular conditions the main mechanisms are bound-free (photoionisation) and free-free (inverse Bremsstrahlung) transitions. To determine the appropriate ratios the code uses the sample charge state which is obtained from tabulated values via the current electron temperature which for simplicity is assumed to settle on a timescale shorter than the pulse durations. The simulations confirm that the soft x-ray FEL pulse heats the sample more homogeneously than the infrared laser pulse. We are currently investigating the effect of inhomogeneous heating of the liquid hydrogen jet by the infrared laser leading to variations of the electron temperature and density. Initial analysis indicates that the use of the plasmon peak to estimate the plasma parameters provides good accuracy [28].

For the experimental conditions of these experiments we obtain the following relation for the scattering parameter

$$\alpha = \frac{e\sqrt{n_e}}{K\sqrt{\varepsilon_0 k_B T_e}} \cong 20.4 \times \frac{\sqrt{n_e [10^{22} \text{ cm}^{-3}]}}{\sqrt{T_e [\text{eV}]}} , \quad (1)$$

that is shown graphically in Fig. 2. One finds that in the case of liquid hydrogen scattering occurs in the collective regime for all densities between the liquid ($n_e = 4.2 \times 10^{22} \text{ cm}^{-3}$) and expanded densities of below $n_e \approx 10^{20} \text{ cm}^{-3}$ if one assumes that the electron temperatures do not exceed a few eV. For solids ($n_e \approx 10^{23} \text{ cm}^{-3}$) temperatures can be considerably higher still fulfilling the condition for collective scattering. Using 60 μJ FEL pulses at the sample and scattering from a liquid hydrogen jet of $\sim 20 \mu\text{m}$ diameter we are able to obtain significant scattering from a single pulse (compare Fig. 3). These are ideal conditions for this experiment, as they allow measuring the beam properties of FEL beam on a pulse-by-pulse basis. Unfortunately we could not operate the whole experiment in this mode but had to collect scattering data from laser-heated hydrogen plasma using integration over several hundred pulses. During integration both FEL pulse energy and mean density of the liquid jet were fluctuating. In this mode signal was collected over a predefined time and in parallel the mean FEL pulse energies and the spectral distribution were measured for normalization of the scattering data. In order to normalize the fluctuations of the liquid jet a monitoring signal is needed which is linear with respect to the scattering probability of the FEL beam. Two approaches were attempted: First, one can use the transmitted beam intensity to determine the absorbed beam fraction. This method was shown to be unfeasible since a detector for the transmitted beam gets overloaded with the infrared pump laser beam. Second, one can use a fast, pulse resolved scattering signal. Both light scattering and particle emission have been analyzed using a multi-channel plate detector mounted for this purpose. The normalization using this signal works, in principle, providing sufficient counting statistic can be acquired. As this is not always the case, the error margin can be relatively high.

Providing fresh sample volumes is a prerequisite for integrating experiments. Using the liquid jet with an expansion speed ~ 60 m/s we calculate that fresh sample volumes are safely achieved for FEL repetition rates up to 1 MHz. Using the femtosecond infrared laser for pumping the system experiments were carried out using the 5 Hz repetition rate. In the solid experiments the sample was continuously translated using the 5 Hz repetition rate of the FEL.

4. Results and discussion

The results of soft inelastic x-ray scattering experiments from liquid hydrogen can be separated into two areas. These are: a) the experimental procedures to observe scattering of FEL radiation and detection using efficient spectrometers and b) the time-resolved investigation of the parameters of an infrared laser prepared hydrogen plasma. The experiment itself can be subdivided into (i) the scattering of FEL radiation including the preparation and characterization of the FEL beam and the spectrometer for efficient detection, (ii) the sample preparation and characterization and (iii) the infrared laser heating of the hydrogen plasmas.

For soft x-rays absorption is considerable even in the case of the reduced density of liquid hydrogen. The energy absorbed from the focused FEL radiation will therefore itself lead to strong excitation of the sample that eventually leads to plasma formation. The amount to which a transient change of the sample affects the inelastic scattering, a process called self-scattering, is the issue of a forthcoming publication [29]. The experimental results show that the two parameters of the FEL radiation that are most critical to the success of these experiments are the pulse energy and the bandwidth. It is

found that a pulse energy of order 30 – 50 μJ is needed to obtain a significant amount of scattered signal using single pulses or very short (few 10 seconds) integration times. In addition, it is crucial that the liquid hydrogen source operates stably throughout the integration time in the high average density liquid jet mode. Short integration times can be still acceptable as the stabilization of parameters can work on this time scale. Further, it is extremely critical for the observation of plasmon peaks that the bandwidth of incident FEL radiation be sufficiently small and the spectral resolution of the spectrometer be sufficiently high. As shown in Ref. [7] the shift of the plasmon signal with respect to the elastic scattering is in the order of only a few eV. The broadening of the spectroscopic data due to the incident bandwidth or by spectrometer resolution would wash out the experimental results. A total spectral resolution better than 1% is therefore required for the conditions of these experiments. Figure 4 shows experimental data obtained from liquid hydrogen. Spectra shown are for the cases of infrared laser arriving 3 ps before and 3 ps after the FEL pulse and integrating over 9000 pulses at 5 Hz. Since the two spectra do not differ significantly we subtract them after background subtraction and normalization to the same value. The resulting difference signal yields a small signal. Also shown are the corresponding spectra of the incident FEL radiation, measured at $\theta = 0$ degrees and using the same integration times as for the scattered spectra. After normalization the difference profile of the incident FEL radiation shows features similar to the difference profile of scattered radiation. In this case we conclude that likely fluctuations of the spectral composition of FEL radiation over the integration period of the measurement are responsible for the difference profiles. This result demonstrates the crucial importance of a simultaneous measurement of the spectral composition of FEL radiation and the requirement that the FEL must operate under very stable conditions in

order to enable experiments of this kind. Comparing the peak width of the scattered radiation and the measured FEL spectrum we observe a clear broadening in the scattered spectrum. Whether this broadening is due to inelastic scattering near the elastic peak or is due to source broadening requires still further clarification. The oscillations of the signal away from the scattering peak is reproducible and is due to density modulations of the 200 nm Zr filter for visible light suppression inside the spectrometer.

The preparation of the liquid hydrogen jet needs to include the characterization of its correct alignment with respect to the cross-section of FEL beam, optical laser and spectrometer axis and of the mean density of the jet during the measurement. These measurements are critical for determining the absolute scattering intensity, e.g. in investigations of the FEL intensity dependence of the amount of self-scattering. We further note that the fluctuations of the infrared laser pulse energy and beam pointing need to be monitored to ensure identical plasma conditions in these highly repetitive measurements.

These experiments are challenging in that FEL, liquid hydrogen jet, and infrared laser have to operate simultaneously with specified parameters otherwise the experimental outcome will be compromised. Integration over many FEL pulses introduces uncertainty and broadening of experimental parameters which are crucial to the observation of results for the plasma parameters. On the other hand, the operation of FLASH at high pulse energies ($>30 \mu\text{J}$) has shown the feasibility of carrying out these measurements using single pulse spectra. In this case the accumulation of a significant amount of spectral data with good resolution can be achieved by sorting data according to simultaneously

measured FEL and infrared laser data. Also, dependencies related to FEL intensity or sample variation can be followed using this procedure.

The impact of the FEL pulse on solid samples was investigated by observation of emission radiation using the same experimental setup. The deposited energy leads to the formation of a plasma exhibiting emission characteristics that are fundamentally different from those of a plasma created by an infrared laser of similar intensity. Figure 5 shows a comparison of the emission spectra of a bulk aluminum sample for FEL and infrared laser irradiation. Following soft x-ray excitation one observes, in addition to the elastically scattered soft x-ray radiation, a strong difference of ion line emission and continuum radiation indicating the different plasma state the aluminum has turned into after soft x-ray irradiation. In contrast to the elastic and near-elastic scattering the continuum, the ion line emission can occur at delayed times after impact of the FEL pulse. The spectra shown in Fig. 5 integrate over timescales up to ns in which the plasmas may emit. Detailed results of this characterization of FEL created warm dense Al plasmas using soft x-ray emission spectra have been published separately [14]. In order to achieve high statistical accuracy during these measurements they are carried out integrating over a large number of FEL pulses. The variation of FEL pulse energy and the modification of sample conditions related to the high repetition rate of the FEL pulses require further investigation using single pulse spectra.

5. Conclusions

The experimental procedures for soft x-ray inelastic scattering experiments on cryogenic liquid hydrogen jets using FEL radiation have been developed. The observation of single-

pulse scattering spectra using efficient spectrometers is found to be possible. First results have been obtained for the time-resolved investigation of the parameters of an infrared laser prepared hydrogen plasma. The interaction of the FEL pulse with the sample leading to plasma heating, thereby changing the measured state, is still under investigation. The experiments reported here were the first of their kind carried out using FEL radiation. The results of these experiments indicate the requirements for future experimental campaigns with respect to diagnostics of FEL and infrared laser radiation as well as sample characterization. Scattering data has been collected using an integrating 5 Hz mode for laser heated hydrogen plasmas varying the time delay between excitation and probing. These results will be subject of future work. Most importantly we found that single pulse scattering on liquid hydrogen gives significant scattering signal for a detailed study of plasma conditions. Applying single pulse data collection offers the advantage of reducing statistical errors and experimental uncertainty introduced by integration over varying parameters.

For solid aluminum samples the absorption of intense soft x-ray FEL radiation and subsequent formation of warm dense plasmas has been investigated by observation of the time-integrated emission spectrum which differs strongly from the spectrum after heating using infrared lasers.

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Figure captions:

Fig. 1 (colour in electronic-; b/w in print-version)

Single-pulse spectra indicating the pulse-to-pulse fluctuation of the spectral distribution of incident SASE FEL pulses. The bold line corresponds to the spectral distribution averaged over 8 pulses and indicating the incident bandwidth of ~ 2 eV corresponding to ~ 2 % relative bandwidth.

Fig. 2 (colour in electronic-; b/w in print-version)

Phase space representation for electron density n_e and temperature T_e , probing photon energy $\hbar\omega = 92\text{eV}$ and scattering angle $\theta = 90\text{deg}$. Lines for $\alpha = 0.5, 1, 2$ and $\Gamma = 1$ have been calculated according to the relations in the text and in equation (1). Grey areas indicate the expected parameter regimes for hydrogen and Al plasmas investigated. Densities of cold hydrogen and aluminum are indicated.

Fig. 3 (colour in electronic-; b/w in print-version)

Scattering signal from a single FEL pulse of $60\ \mu\text{J}$ energy at the sample obtained at the CCD of the HiTraX spectrometer [20] after background subtraction. 500 counts in the maximum correspond to a very high S/N ratio that is well beyond 50.

Fig. 4 (colour in electronic-; b/w in print-version)

Soft x-ray Thomson scattering spectra near $92\ \text{eV}$ from hydrogen integrated over 9000 FEL pulses. Scattering spectra for the cases that the $800\ \text{nm}$ infrared laser hit the liquid hydrogen $3\ \text{ps}$ prior (dots, broken line) and $3\ \text{ps}$ after (crosses, dotted line) the FEL pulse are shown. Difference scattering spectra (error symbols, line) have been obtained from normalized scatter data after background subtraction. The corresponding FEL spectra and their difference are shown as dash-dotted lines.

Fig. 5 (colour in electronic-; b/w in print-version)

The upper curves shows an unfiltered soft x-ray spectrum obtained from bulk aluminium following impact of $92\ \text{eV}$ FEL radiation and the lower curve the spectrum for $1.6\ \text{eV}$

infrared laser radiation using a 200 nm Zr filter. The intensities were $4 \times 10^{14} \text{ Wcm}^{-2}$ and $4 \times 10^{15} \text{ Wcm}^{-2}$, respectively.

Figure 1

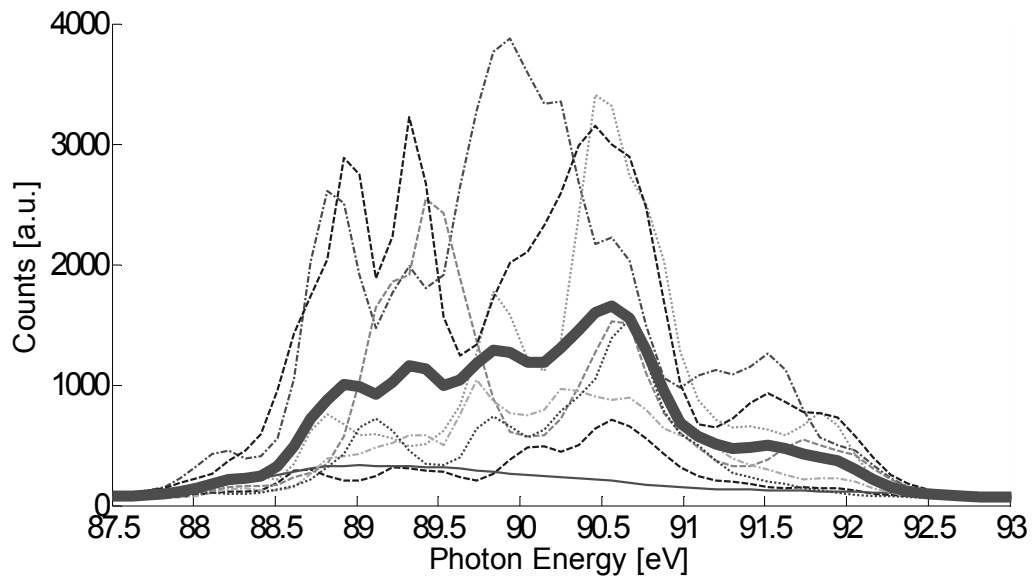
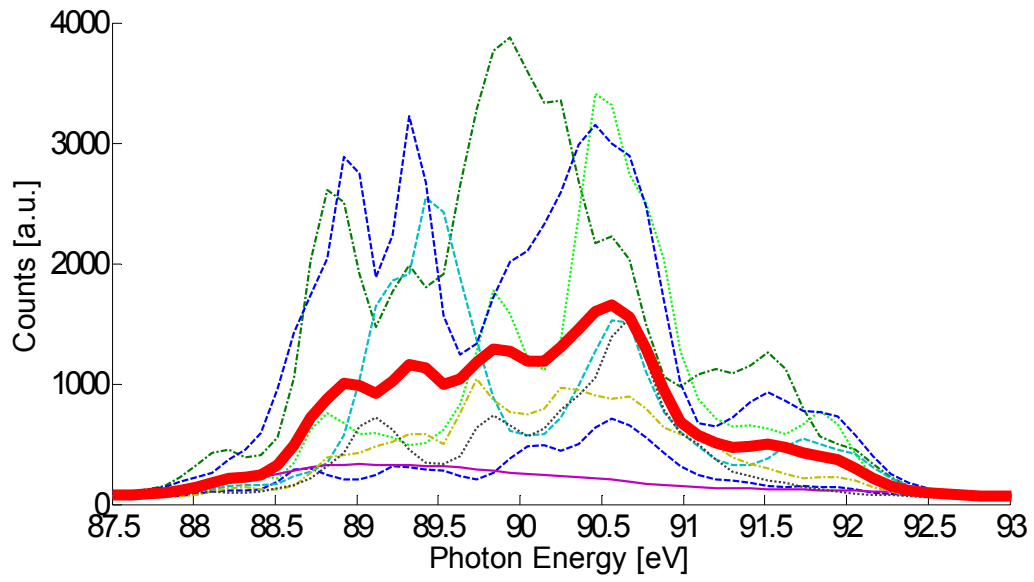


Figure 2

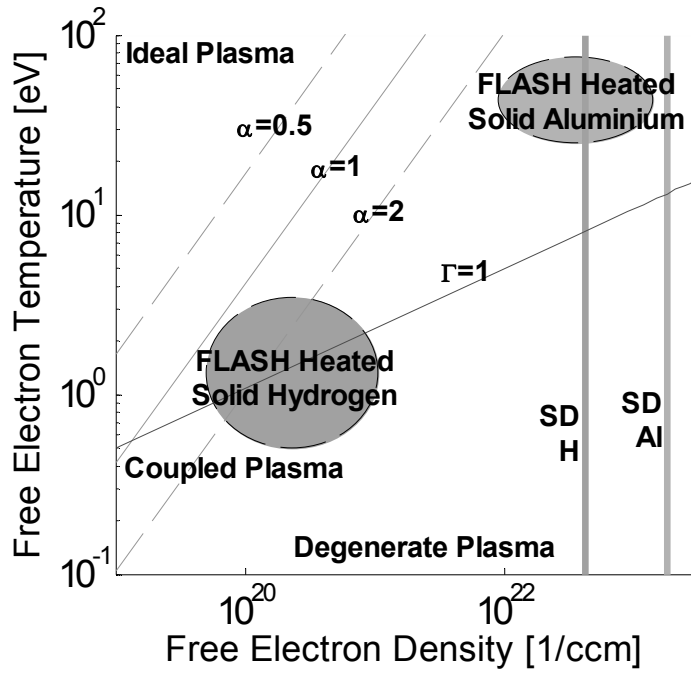
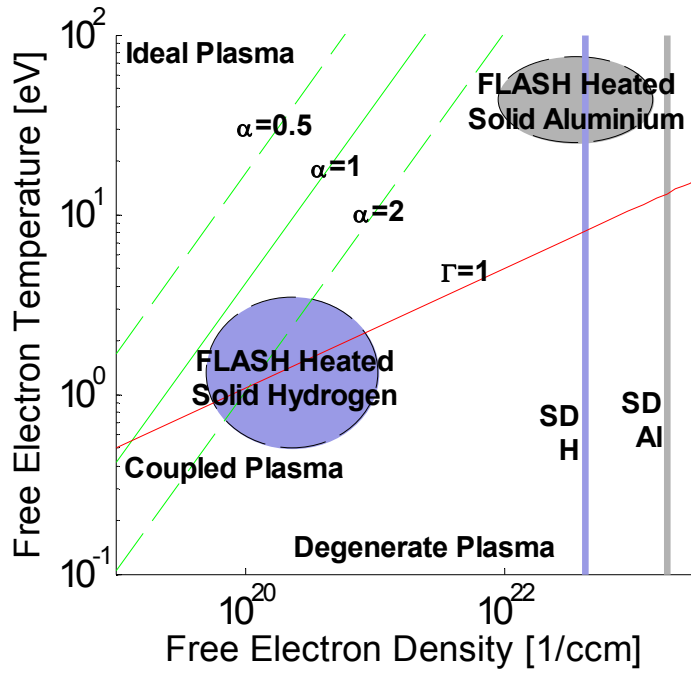


Figure 3

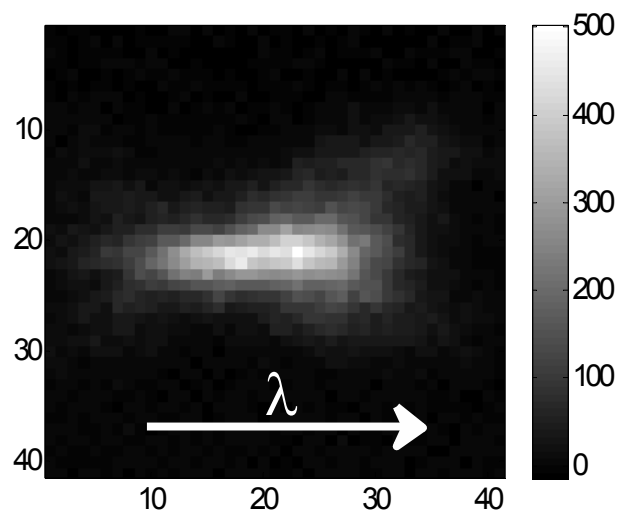
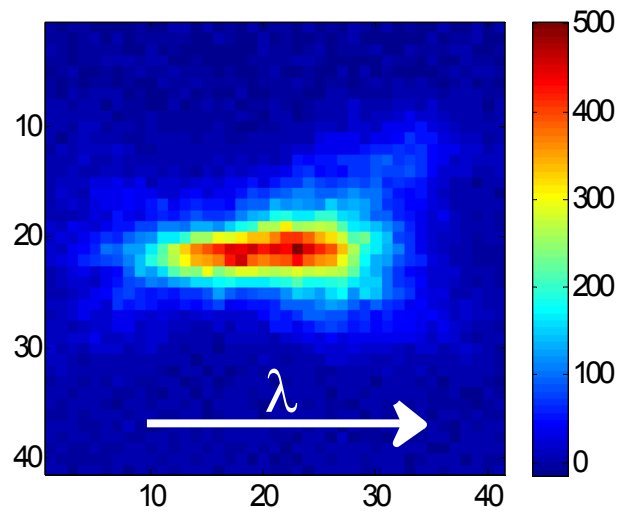


Figure 4

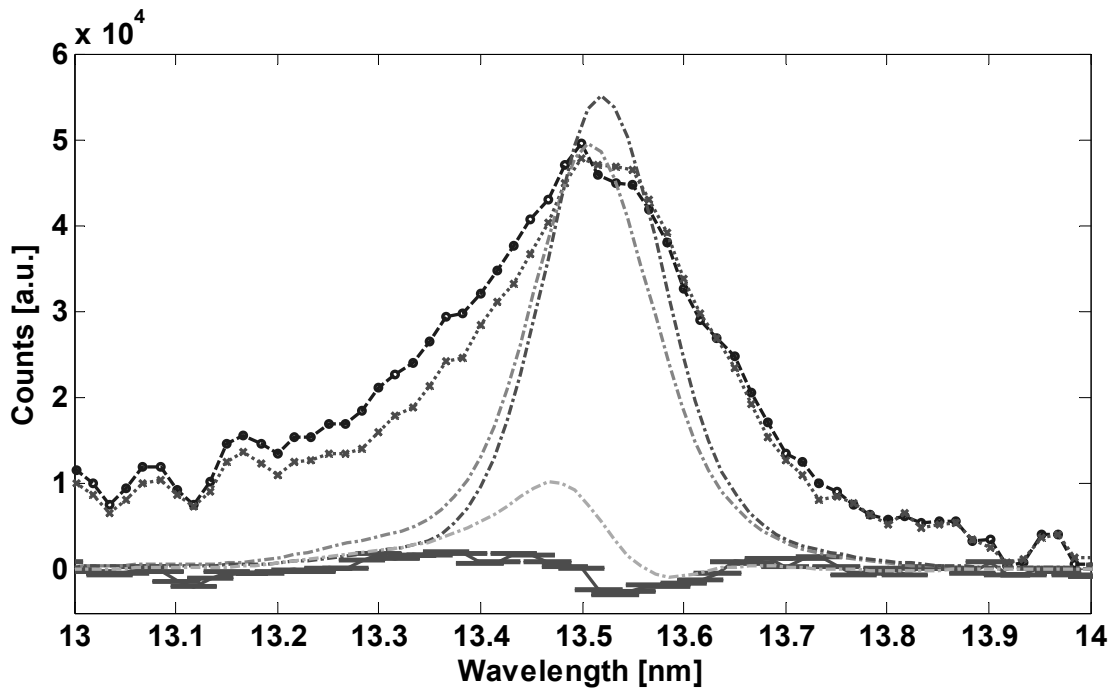
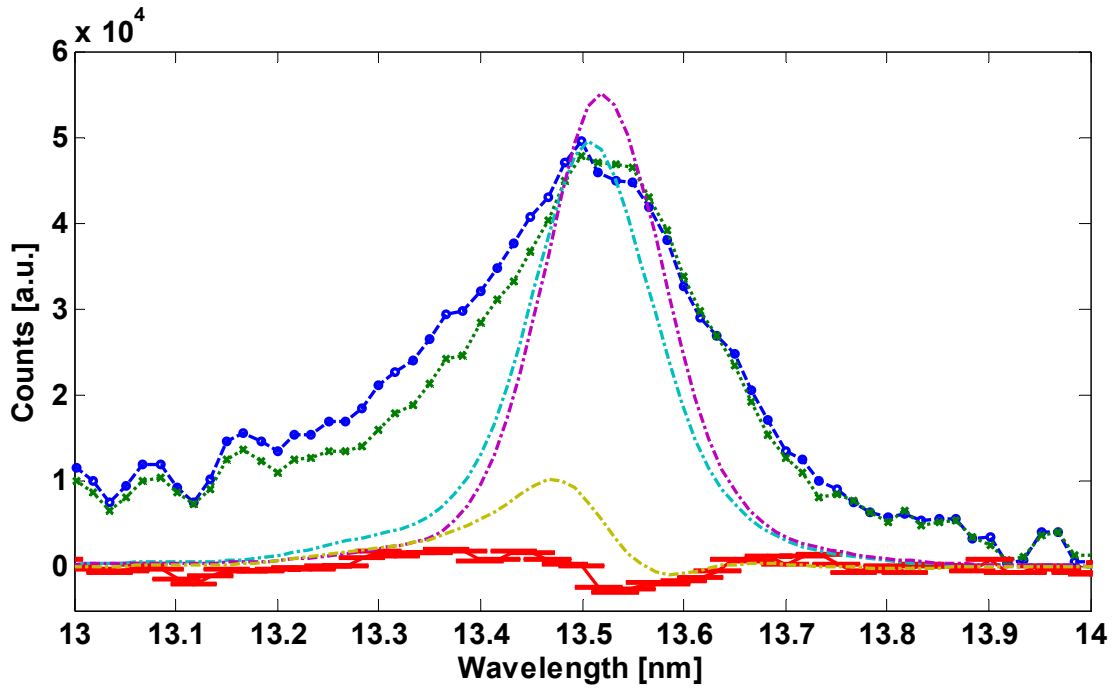


Figure 5

