Probing matter under extreme conditions at FERMI@Elettra: the TIMEX beamline

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ABSTRACT

FERMI@Elettra is a new free-electron-laser (FEL) facility, presently under commissioning, able to generate subpicosecond photon pulses of high intensity in the far ultraviolet and soft X-ray range (λ=100-20 nm for the present FEL1 source, extended in future to 4 nm with the FEL2 source). Here we briefly describe the present status of the TIMEX end-station, devoted to perform experiments on condensed matter under extreme conditions. The layout of the end-station, presently in the final stages of construction, is reported showing the details of the optics and sample environment. The potential for transmission, reflection, scattering, as well as pump-and-probe experiments is discussed taking into account that FEL pulses can heat thin samples up to the warm dense matter (WDM) regime. The calculated deposited energy in selected elemental films, including saturation effects, shows that homogeneous heating up to very high temperatures (1-10 eV for the electrons) can be easily reached with a suitable tuning of the energy and focus of the soft x-ray pulses of FERMI@Elettra. The results of the first test of the TIMEX end-station are also reported.

Keywords: free electron laser, extreme conditions, warm dense matter

1. INTRODUCTION

The TIMEX beamline is conceived to exploit the unique opportunities offered by the FERMI@Elettra free electron laser (FEL) source[1,2] for studies of condensed matter under extreme conditions.[3,4] In the TIMEX scientific program, the FEL beam will be used as a pump and/or probe for ultrafast (reflectivity, transmission, scattering) studies mainly concerning (i) generation of warm-dense-matter (WDM), (ii) transitions occurring in stable, metastable and excited states under transient high-temperature/pressure conditions and (iii) ultrafast surface/bulk transitions and ablation phenomena.

The FERMI@Elettra FEL generates subpicosecond pulses of selected energies and photon wavelengths (100-4 nm range), containing up to 10^{14} photons in a single-shot, which can be used to raise the temperature of specimens by exciting a large fraction of the electrons. A typical sample equilibrates its temperatures within a few picoseconds and can reach very high temperatures (up to 10^3-10^5 K) still maintaining typical densities of condensed matter. The FEL high single-shot deposited energy in condensed matter can pump selected specimens in the WDM regime, where the thermal energy is comparable to that of the interatomic potential.[5] This state of matter is poorly known and its knowledge is of basic interest because such disordered states are those found in the interior of large planets and in stars and can be produced in devices for plasma production and inertial confinement fusion. The intensity of the FEL pulses was recently used for creating and investigating matter

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in the WDM regime at the fourth-generation light source FLASH (Hamburg)[6,7] and is part of the scientific program of the LCLS (MEC beamline) x-ray FEL (Stanford).

Probing condensed matter under extreme transient conditions using FEL radiation is particularly promising also because it extends the ultrafast techniques already available using optical lasers to homogeneously bulk-heated specimens, opening new perspectives to study the dynamics of transitions (melting for instance) in ordered and disordered condensed matter. Moreover, ultrafast experiments give access to presently unreachable states of matter (no man's land) because of their extremely fast transition rates. We planned to use several techniques for probing matter under these extreme conditions at the TIMEX beamline, including absorption spectroscopy, reflectivity and pump-and-probe experiments.

In this contribution we describe the technical and scientific case of the TIMEX beamline, illustrating the layout of the instrument (Sec. 2), the optics and expected performances (Sec. 3), and the present status of the end-station (Sec. 4). Perspectives and concluding remarks are reported in Sec. 5 while the involved institutions are acknowledged at the end of the paper.

2. LAYOUT OF THE TIMEX END-STATION

FERMI@Elettra is a 4^{th} generation light source user facility with a high brilliance and short pulse length operating at Sincrotrone Trieste since early 2011. The High Gain Harmonic Generation (HGHG) scheme[8] employing multiple undulators is conceived to use the initial seed signal (pulsed laser) providing ideally transform-limited and spatially coherent radiation pulses in the far ultraviolet/soft x-ray range. Duration, bandwidth and wavelength of the output radiation is expected to be tunable within times of minutes.

The first undulator chain (FEL1)[1] has been completed in late 2010 and is presently under commissioning until reaching the expected final performances. FEL1 covers the wavelength range from 20 nm to 100 nm (down to 6.7 nm by using third harmonic) with more than 10^{13} photons for each subpicosecond pulse. During the first runs FEL1 operated in the 43-65 nm range with an intensity reaching 10^{10}-10^{11} photons/pulse and durations estimated in the 0.2 ps range (compatible with expected performances during the first days). FEL2 is under construction and is expected to provide radiation from 4 nm to 20 nm (down to 1.3 nm in third harmonics), with typical pulse width in the 40-100 fs range and up to $\sim 10^{13}$ photons per pulse (see[2,4] and ref. therein).

The FEL pulses are delivered to the beamlines through a dedicated system (PADReS[9]) for diagnostic and intensity tuning, presently under commissioning and test. A suitable optics has been designed for the TIMEX end-station providing unique beam-shaping capabilities for obtaining a 3-50 μm spots with the desired energy (and fluence) deposited on the sample (see next Section and ref.[4]). We are also developing a proper diagnostics for the temperature reached by the sample after the pump pulse, as described in previous papers.[4,10]

As shown in Fig. 1 the beamline design is conceptually simple and includes a delay line (30 ps), a plane mirror with active piezo benders (under construction), an elliptic focusing mirror with focus at 1.4 m at the sample position inside the main UHV (Ultra-High-Vacuum) TIMEX chamber (see side and top views in Fig. 1). All optical elements are presently under construction. Various devices to adjust the intensity of the pulse and to align the beam, like insertable filters and active or passive screens, can be used along the beamline. The sample environment (see top view on the right of Fig. 1) has been kept quite flexible and can accommodate various possible configurations for single-shot experiments including simple far-UV and soft x-ray absorption/reflection, and pump and probe experiments using as a probe either an optical laser or the FEL pulse (and its harmonics). The optical laser can be used as a pump too.

The sample is installed on a motorized sample manipulator stage working in the main TIMEX chamber. This 5-axis motorized manipulator is conceived for single-shot measurements at 10-100 Hz rate, allowing precise alignment of the sample in the interaction region with pump and probe ultrashort pulses. Diagnostic for initial experiments include a long-distance (LD) microscope for fine micrometric alignments and an infra-red pyrometer,[10] but further space is left for additional instrumentation. The main chamber is also equipped with a set of detectors, typically Si diodes or bolometers for measurements of direct photon transmission and reflection. Space is left also for the possible use of an ultrafast streak camera for which we planned to test dynamic ranges and time-resolution. The sample chamber will be fully interfaced through a translation stage with a sample preparation chamber (already commissioned and tested) for transfers of samples without breaking the vacuum

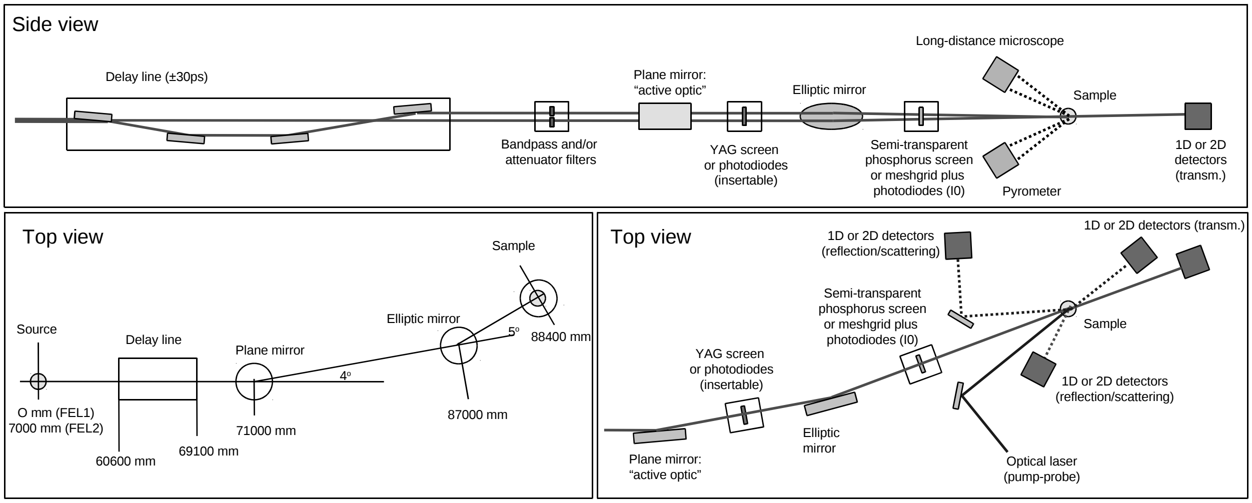

Figure 1. Sketch of the TIMEX end-station under commissioning at the FERMI@Elettra FEL facility. The side view shows all the optical elements up to the sample chamber, most of them presently under construction. In the top view on the left side of the figure we show the main components of the end-station along with their distances from the sources. In the top view, right side, we show the details of the focusing and aligning devices and the detectors used for reflection and transmission measurements using both the FEL and a probe pulse of an optical laser.

and for surface-science characterizations of fresh specimens before one-shot experiments. Further developments are also related to the installation of a table-top supercoontinuum probe system[3, 11] for collecting ultrafast optical absorption and reflectivity data in a wide range of wavelengths in single-shot pump-and-probe experiments (see Fig. 1, right top view). Fast CCD cameras and diode array detectors will be used for detection of optical ultrashort pulses within this pump-and-probe scheme.

3. OPTICS AND EXPECTED PERFORMANCES

The TIMEX end-station is conceived to takes advantage of a focused FEL beam with micrometric size, ensuring the necessary photon fluence for high temperature experiments up to the WDM regime (1-10 eV typical electron temperature).

To achieve this goal for both FEL1 and FEL2 (see ref.[4]), the basic design of the optics of the beamline consists of a plane and a focusing ellipsoidal mirror (see Fig.1). The plane optics will be placed inside the switching chamber in common with the Low Density Matter (LDM) beamline and the Diffraction and Projection Imaging (DiProI) beamline while the focusing mirror will be placed 15 meter downstream with a focal distance of 1.4 meter. Our previous studies on the expected temperature profile, as a result of a FEL pulse interaction at the sample surface,[10] show that knowledge and mastering of the pulse shape are important pre-requisites for obtaining controlled and reliable initial temperature conditions. A flat-top distribution is expected to provide a uniform deposited intensity (and corresponding initial temperature) at focus, while the existence of tails would make it possible a direct measurement of temperature raising with an infra-red pyrometer. Thus, the possibility of modifying the expected shape of the FEL pulse is an important tool for adjusting the deposited energy to the selected level of intensity and spatial homogeneity.

For obtaining almost flat-top spatial distributions at focus, a mirror with unusual shapes has to be used. For this reason the plane mirror was replaced with an active optics that consists of a 400 mm × 40 mm × 10 mm substrate with 15 piezo-actuators glued below the substrate[4] and controlled with an in-house developed power supply (HVPS MAS-TER)[*]. Of course this mirror can also be adjusted to be a simple plane mirror with no

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[*] refer to the webpage http://ilo.elettra.trieste.it/index.php?page=_layout_prodotto&id=122&lang=en

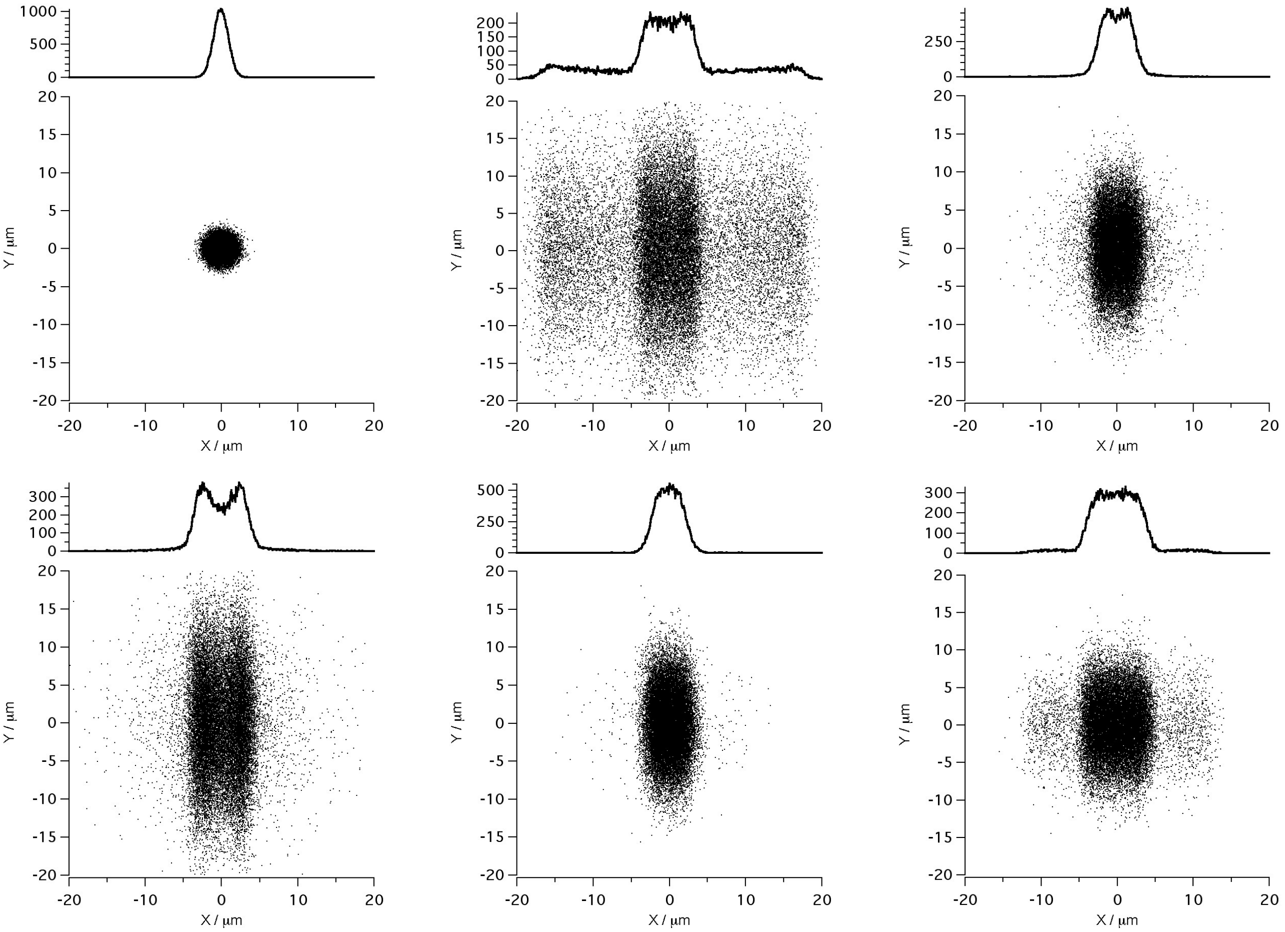

Figure 2. Ray-tracing results of the expected spots at focus with the relative mirror profiles for a wavelength of 5 nm and a divergence of 7.5 μrad. Figures on the upper row from left to right: i) ideal plane mirror (Gaussian distribution); ii) γ= 4 cm, α= 8, A= 100 nm (flat-top distribution with tails); iii) γ= 8 cm, α= 2 A= 150 nm (flat-top/double peak). Bottom, from left to right: i) γ= 8 cm, α= 2, A= 250nm (double peak); (ii) γ= 8cm, α= 4, A= 150 nm (flat-top distribution), (iii) γ= 8 cm, α= 8, A= 150 nm (larger flat-top distr.). The mirror profile is assumed to be Lorentzian-like (see text) along the x direction (x_0=0 cm).

extra-curvatures. We have found that by modifying slightly the mirror profile with simple analytical functions (Lorentzian-like, for example) we can obtain spots with various shapes of interest. We have thus performed ray-tracing simulations using a Lorentzian-like function for the mirror profile:

$$L_\alpha(x)\,[\gamma, A, x_0] = A\frac{\gamma^{\alpha-1}}{\pi\left[(x-x_0)^\alpha+\gamma^\alpha\right]} \tag{1}$$

where γ is the half width at half maximum (HWHM), A is the amplitude, x_0 is the center of symmetry and α is an even number. Metrological test have been successfully completed and we have shown that the ideal needed shapes can be easily reached with the active optics. The results of selected mirror profiles on the focus shapes are reported in Fig. 2.

As mentioned above, the FEL pulses generated by FEL1 or FEL2 sources are expected to interact strongly with condensed matter giving rise to high levels of deposited energy when proper focusing is achieved as shown in Fig. 2. The amount of deposited energy depends on various factors related to the source, optics (number of photons, photon energy, spot dimensions) and target (material, thickness). In Fig. 3 we show the results of a simple calculation of deposited energy for a thin aluminum foil (0.3 μm), using pulse parameters compatible with the FEL1 source and the optics of the TIMEX beamline: photon wavelength $\lambda = 60$ nm, spot 10x10 μm^2, number of photons 10^{11}-10^{14}. Self-standing foils of this uniform total thickness (0.1-0.3 μm) have the suitable robustness and reliability needed in real single-shot experiments.

The dashed curves in Fig. 3 represent the energy deposited through the film using tabulated atomic scattering factors and absorption cross-sections[12] and standard linear absorption with no saturation (Beer-Lambert law). However, due to the intense electromagnetic field energy density (number of photons comparable with the number of available electrons in the interaction volume) strong saturation effects may be expected. At sufficiently high incident intensity, atoms in the ground state of the target become excited at such a rate that there is insufficient time for them to decay back to the ground state, and the absorption subsequently saturates. The solid lines reported in Fig. 3 have been obtained using a simple model for saturation effects for a typical absorption channel represented by the absorption coefficient α_s:

$$dI(x) = -\frac{\alpha_s I(x) dx}{(1 + I(x)/I_{sat})}. \tag{2}$$

In Eq. 2 the number of photon absorbed (lost) per unit area $dI(x)$ at depth x inside the target is proportional to the incoming flux $I(x)$ and to the absorption coefficient as usual, but only for intensities well below a saturation threshold I_{sat}. Different absorption channels (electrons in different ground states) can show different saturation levels for a given photon excitation energy. The saturation threshold used to calculate the curves shown in Fig. 3 has been chosen in order to reproduce the saturable x-ray absorption observed in a ultra-thin Al foil by Nagler et al.[6] The saturable channel is considered to be 80% of the total cross section (20% of residual absorbance) and the threshold density of photons is approximated by $N_{sat} = Kn_{el}$ where $K \sim 1$ and n_{el} is the density of excitable electrons. The resulting electron temperature T_e, shown also in Fig. 3 is estimated by equating the energy deposited per unit volume with the sum of the internal and kinetic energies of the sample.[5] Depending on the incoming number of photons, electron temperatures in the range 0.1 to 5 eV can be obtained with the given conditions. It is important to remark that saturation provides an excellent tool to produce quasi-isotherm bulk heating especially for high fluence,[†] although it obviously limits the average expected temperature.

The photon energy tunability of the FEL1 and FEL2 sources represents an interesting additional tool for adjusting the deposited energy into selected specimens at the TIMEX beamline. In fact, the natural energy dependence of the absorption cross-section modulates the deposited energy and the resulting temperature of the sample. We have considered a set of large-size thin foils that are actually available and can be used for routine shot-to-shot experiments (C, Al, Si, Ge). In Fig. 4 we report the deposited energy in different thin film materials as a function of the photon energy in the range useful for FEL1 and FEL2, for pulse intensities corresponding to 10^{12} photons and focus spot of 10x10 μm^2. The average deposited energy (see solid lines) through the films show different trends according to the particular film under consideration. Film thickness was chosen in a range (0.1-0.3 μm) for which robust and wide-area samples are available for repeated shot-to-shot experiments. Depending on the photon energy, there is a spread on the deposited energy for different depths inside the films (see Fig. 3) and upper and lower bounds are shown in Fig. 4 (dashed). Calculations reported in Fig. 4 were performed accounting for possible saturation effects as discussed above. Looking at Fig. 4 we notice that there are large variations in the deposited energy and spread as a function of photon energy, mainly depending on the absorption cross-section (so on the opening on new electron excitation channels). By a suitable tuning the FEL energy, we see that almost uniform bulk heating can be obtained as shown by the narrow regions limited by the dashed curves (light green, color on-line). On the other hand, extremely inhomogeneous heating can be obtained when there is a rise of the cross section and most of the energy is deposited near the surface of the thin film as shown by the spread of the dashed curves (pink, color on-line). As shown in Fig. 4, average and uniform electron temperatures T_e in the 1-10 eV range can be easily reached at different energies depending on the nature of the film. In particular, the radiation corresponding to photon energies in the 20-60 eV range (covered by FEL1) is useful for obtaining uniform bulk heating in various thin films, like those of Al, Si, Ge reported in Fig. 4. Films of carbon, instead, are found to be extremely absorbing and therefore uniform heating

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[†] This effect has been noted also by R. W. Lee and communicated privately to one of the authors (A. D. C.)

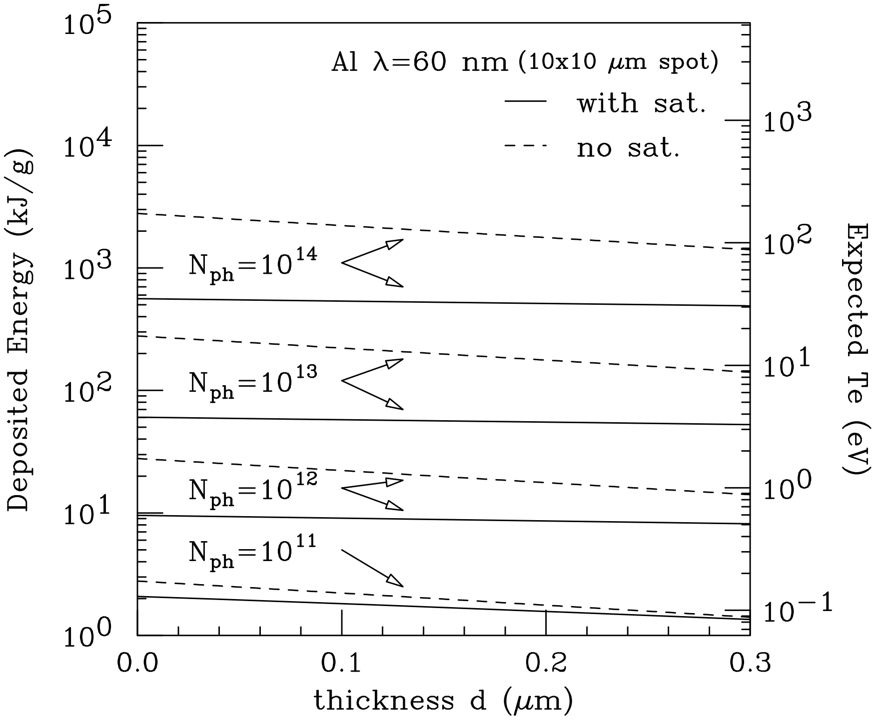

Figure 3. Deposited energy (kJ per gram of substance) as a function of the thickness d for a thin aluminum foil, for different intensities of the FEL pulses (10^{11} to 10^{14}) at a wavelength of 60 nm (spot 10x10 μm^2). The dashed curves are obtained without considering saturation effects due to the intensity of the pulse. The solid curves are obtained using a simple model for saturation. Saturation is expected to provide for high pulse intensities a much more uniform bulk heating (see flatness of the solid curves) and a decreased deposited energy. The expected electron temperature T_e is also shown.

can be obtained only for photon energies higher than 60 eV (films much thinner than 0.1 μm can still be used in the range useful for FEL1).

Finally, it is interesting to remark that the FEL radiation in the soft x-ray range covered by FEL1 (and FEL2) is found to be extremely efficient for obtaining uniform bulk heating of thin films up to the WDM regime. The pulse energy needed for this purpose is generally lower than that needed at higher photon energies, due to the opacity of most materials in the far ultraviolet/soft x-ray ranges (note that in Fig. 4 the pulse energy increase of 1 order of magnitude for fixed number of photons).

4. PRESENT STATUS OF THE END-STATION

The end-station has been tested very recently (March 2011) using the first available beamtime at FERMI@Elettra (λ = 65 nm). The experimental hall has been made available for the installation of the instrumentation only in the last few months so those first runs have been carried out using only part of the available devices (the focusing optics is still under construction). In particular, the TIMEX main sample chamber (see Fig. 5) has been installed just after a temporary focusing mirror placed in a different chamber, providing a spot size of about 100 μ at sample position. The position of these two chambers roughly corresponds to the plane mirror position in Fig. 1.

In Fig. 5 (picture on the left) we can see the main components of the TIMEX end-station installed in the Fermi experimental hall. The Fermi pulses proceed into the beamline for several meters up to the mirror chamber (hidden in the figure) and then into the main TIMEX chamber (the yellow dashed line is a guide for the eye). Several insertable fluorescent screens and filters have been positioned along the beamline in order to align the beam, as well as safety shutters and valves controlled by the beamline control software. The software has been

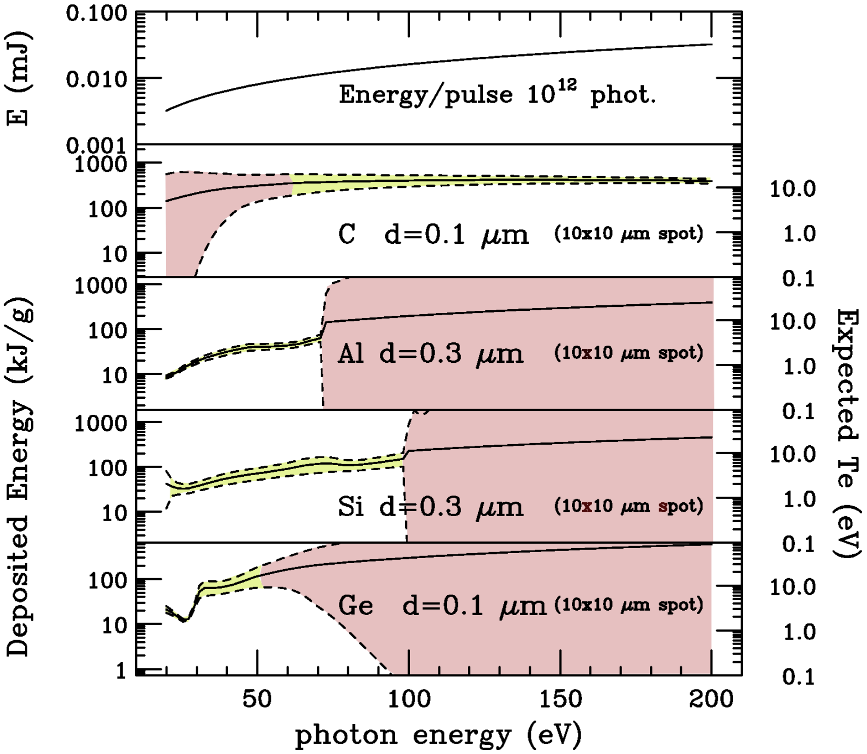

Figure 4. In the lower four panels we report the deposited energy (kJ per gram of substance) in different thin film materials as a function of the photon energy for a pulse containing 10^{12} photons (spot 10x10 μm^2). The deposited energy (solid curves) has been calculated accounting for possible saturation effects and show different trends according to the particular foil under consideration. The dashed curves represent the limits for the spread of deposited energy inside the films, so uniform bulk heating is obtained when the dashed curves define a narrow region (light green, color on-line). On the other hand, when most of the energy is deposited near the surface of the thin film, an extremely inhomogeneous heating is obtained (pink regions, color on-line). The expected electron temperature T_e is also shown on the right axis, showing that average temperatures in the 1-10 eV range are easily reached depending on the energy and thin foil material. The upper panel reports the pulse energy (mJ) as a function of the photon energy (eV) obviously changing of 1 order of magnitude from 20 to 200 eV. The FEL1 source (12-62 eV) turns out to be extremely efficient for obtaining uniform bulk heating of various materials (for example Al, Si, Ge shown in the picture).

designed in order to control the 5-axis manipulator and read the various detectors (photodiodes, bolometers) to be used for absorption or reflectance experiments. In Fig. 5 we can notice the manipulator and the detector used for transmission measurements.

The first runs were mainly dedicated to a careful alignment of the pulses along the beamline. Due to the relative weak intensity of the FEL pulses in this first stage, no damage or substantial heating of targets were expected so the main task of this first run was basically limited to the measurement of the beam intensity in the TIMEX experimental chamber. The total amount of radiation intensity coming in the TIMEX experimental chamber was measured by using a photodiode (SXUV100, IRD-inc.) placed along the beam path. The signal from the diode was collected and visualized by an oscilloscope (SDA11000, LeCroy) on a shot-to-shot basis. Representative traces are reported in Fig. 5 (right). One of the sources of uncertainty has been the presence of a strong contribution associated with the radiation of the laser seed. We have thus used a 0.3 μm Al film as a filter (transmission $\sim$ 20 % at the FEL photon energy, taking into account the presence of oxide layers at the surface). Data were collected with and without the Al filter inserted along the beam path. With the Al filter, no contributions from the seed laser (photon energy 4.8 eV) were observed. Without the filter the total radiation intensity is still large enough for inducing saturation effects on the detector, as can be readily appreciated by inspecting Fig. 5 (right). In this case the responsivity of the photodiode depends on the radiation intensity. The total photo-generated charge collected at each shot was simply determined as the time integral of the oscilloscope traces (divided by the resistance of 50 Ohm). Typical values of 0.1-1 ($>$ 10) nC was observed with (without) the Al filter inserted along the beam path. According to manufacturer specifications concerning the detector responsivity, we estimated a (peak) FEL energy of about 0.18 μJ after the Al filter, which, taking into account the throughput of the filter and of the photon transport system, reflects into a FEL energy of $\sim$ 1 $\mu J/shot$ at the source, reasonable in this early stage of the commissioning of the FERMI@Elettra source. Even though a straight measurement of the radiation intensity without the Al filter is prevented by saturation effects, we can assess that the contribution of the seed laser radiation to the total signal is as large as the FEL one. A rough estimate of the detector responsivity as a function of pulse energy in presence of saturation effects was done using a femtosecond IR laser source (photon energy 1.5 eV). Within the hypothesis that the relative variation of detector responsivity as a function of pulse energy observed in the IR range can be applied in the UV range, the contribution of the seed laser can be quantified in $\sim 1 - 2\mu J/shot$.

5. CONCLUSIONS AND PERSPECTIVES

In this contribution we described the current developments associated with the construction of the TIMEX beamline, operating at the FERMI@Elettra FEL facility. The design of the beamline, the optics and the expected performances are discussed, with a special emphasis on the possibility of creating and studying condensed matter under (transient) extremely high temperature conditions.

Our estimates indicate that electron temperatures in the range 1-10 eV can be obtained in several selected materials by using the FEL1 radiation (first FERMI@Elettra FEL presently under commissioning) under normal operating conditions. We have found that the FEL radiation in the soft x-ray range covered by FEL1 (and also FEL2) is extremely efficient for obtaining uniform bulk heating of thin films up to the WDM regime. Obtaining high-energy FEL pulses in the low photon energy range is much easier with current FEL technologies and this encourage the use of the TIMEX beamline and FEL1 (and FEL2 in the future) pulses for unique measurements under extreme high-temperature conditions.

The TIMEX end-station has been tested during the first days of operation of the FEL1 source. We have measured a photon fluence compatible with the expected performances of the source at this stage of commissioning.

These initial tests make us confident about the success of the forthcoming experiments on condensed matter under extreme conditions to be performed at the TIMEX beamline. Initial experiments will be oriented to collect absorption and reflectivity data at fixed energy of pure materials interacting with the FEL pulses while ultrafast pump-and-probe experiments will follow. Owing to the FEL photon energy tunability, several other possibilities are open also to experiments using the 1^{st} or higher harmonics, including near-edge absorption spectra at extreme conditions and reflectivity spectra in the soft x-ray regime.

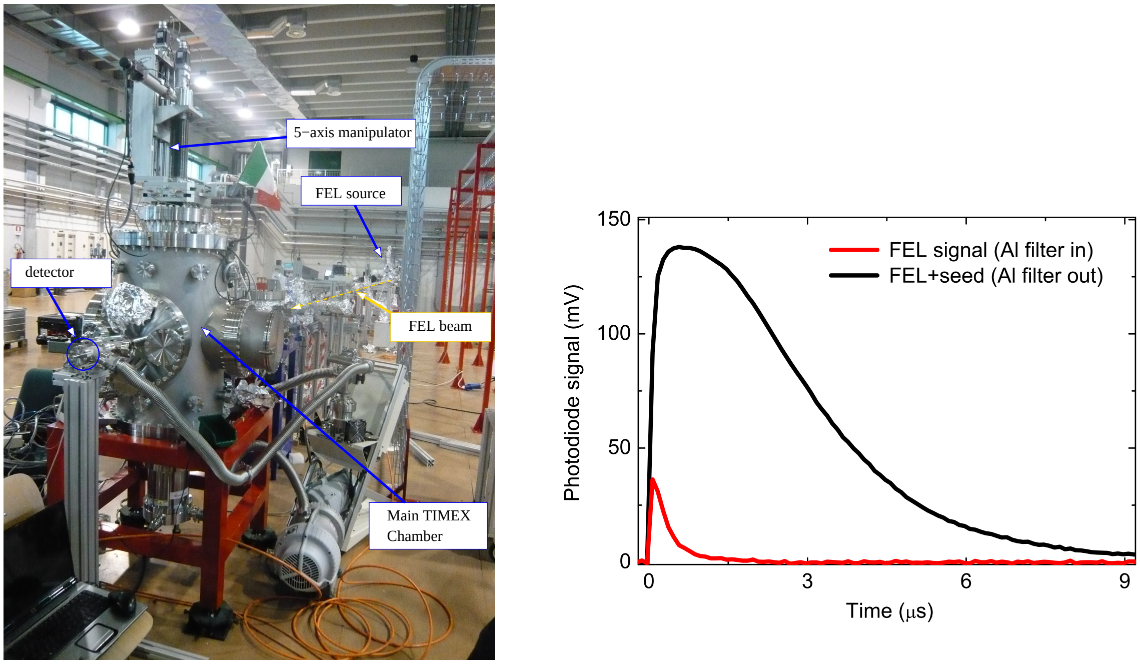

Figure 5. Left figure: first temporary installation of the TIMEX chamber at the exit of the FEL1 source. The FEL beam (yellow dashed line, guide for the eye) is aligned up to the main TIMEX chamber, where the sample position can be controlled with a 5-axis motorized manipulator while the transmitted (detector shown in figure) and reflected pulses can be measured by the photodiodes. Picture has been taken on 17 March 2011. Right figure: traces of the photodiode signal on the oscilloscope. Red (black) curve (color on-line) is a representative single shot measurement of the total radiation intensity coming in the TIMEX chamber with (without) the Al filter inserted along the beam path. From these data we estimated an overall FEL radiation energy of about 1 $\mu J/shot$ at the source and, most likely, we also observed a non negligible contribution ($\sim 1 - 2\mu J/shot$) coming from the seed laser.

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