Journal of Non-Crystalline Solids 357 (2011) 2641–2647

Contents lists available at ScienceDirect

Journal of Non-Crystalline Solids

journal homepage: www.elsevier.com/ locate/ jnoncrysol

Probing phase transitions under extreme conditions by ultrafast techniques: Advances at the Fermi@Elettra free-electron-laser facility

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ARTICLE INFO

Article history:
Received 7 October 2010
Accepted 19 February 2011
Available online 21 March 2011

Keywords:
Free electron;
Laser high-temperature;
Liquids femto-second experiments;
Warm dense matter

ABSTRACT

Novel possibilities for studying matter under extreme conditions are opened by the forthcoming availability of free electron laser (FEL) facilities generating subpicosecond photon pulses of high intensity in the VUV and X-ray range, which are able to heat thin samples up to the warm dense matter (WDM) regime. Pump-and-probe ultrafast techniques can be used to study the dynamics of phase transitions and characterize the states under extreme and metastable conditions. Ultrafast (10–100 fs) bulk heating is seen as a novel route for accessing extremely high temperature regimes as well as the transition region between low-density and high density fluids, that is presently considered a no man's land in simple liquids and glasses. Here we briefly describe the present status of the TIMEX end-station devoted to those experimental activities at the Fermi@Elettra FEL facility, and some preliminary results obtained in a pilot ultrafast experiment using a laser source as a pump and a supercontinuum probe aimed to characterize the melting process of Silicon.

1. Introduction

The forthcoming availability of free electron laser (FEL) facilities offers a unique opportunity to study states of matter under extreme conditions, obtained by exposing condensed matter to their subpicosecond photon pulses. The expected energy and intensity tunability of these potentially extremely intense pulses are able to pump selected specimens in a wide range of temperatures (up to 10^3-10^5 K) while maintaining densities typical of condensed matter under ambient conditions on a typical timescale of a few picoseconds. In the warm dense matter (WDM) regime, easily reached for intense FEL pulses, the thermal energy is comparable to that of the interatomic potential thus leading to an exotic state consisting of a dense electron plasma strongly coupled to the lattice ions [1]. Although being exotic on earth, 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 in the WDM regime at the fourth-generation light source FLASH (Hamburg) [2,3]. The use of FEL radiation for investigating matter under extreme conditions is particularly promising because it extends the ultrafast techniques already available using optical lasers to homogeneously bulk-heated specimens, opening the way to the study of the dynamics of transitions in ordered and disordered condensed matter. Several techniques can be used to probe matter under these extreme conditions including absorption spectroscopy, reflectivity and pump-and-probe experiments. In particular, pump-and-probe ultrafast experiments are expected to unravel details of the dynamics of phase transitions and characterize the states under extreme and metastable conditions, presently inaccessible. For example, those experiments could shed light on the occurrence of polymorphism and the hypothesis about the existence of a coexistence line and a critical point separating low-density and high-density fluids in a class of substances which include water, C, Ge, Si and their oxides [4–6].

Up to now, several ultrafast optical studies have been carried out on simple crystalline and amorphous system giving important information about melting and recrystallization of various substances (see Ref. [7] for a review on semiconductors). For example, melting of crystalline silicon has been studied since the early times of ultrafast optical spectroscopy (see [8–10] and Ref. therein), while melting, crystallization and re-amorphization were studied for various amorphous semiconductors [11,12].

In this contribution we briefly describe the main characteristics of the TIMEX end-station devoted to time-resolved experiments under extreme conditions at the Fermi@Elettra FEL facility in Trieste (see [13] and Section 2). The potential of the new instrument is illustrated in details, and the preliminary results obtained in pilot ultrafast experiments using a laser source as a pump and a supercontinuum

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doi:10.1016/j.jnoncrysol.2011.02.039

probe are here presented, with the aim of characterizing the melting process of the silicon Si(100) surface.

2. TIMEX end-station design

As outlined above, new possibilities for ultrafast experiments are opened by the development of free-electron-laser facilities. Fermi@Elettra is a 4th generation light source user facility with a high brilliance and short pulse length currently under construction at Sincrotrone Trieste. It is based on a High Gain Harmonic Generation (HGHG) scheme employing multiple undulators up-shifting an initial seed signal (pulsed laser) in a single-pass that will provide an almost ideal transform limited and fully spatial coherent radiation. The duration, bandwidth and wavelength of the output radiation will be tunable. Two undulator's chains will be employed: FEL1 covering the wavelength range from 20 nm to 100 nm (down to 6.7 nm by using third harmonic) and FEL2 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 ~10^{13}–10^{14} photons per pulse (see [14] and Ref. therein).

The FEL pulse is designed to be delivered to the beamlines through a sophisticated system (PADRes, see Ref. [15]) dedicated to the diagnostic and intensity tuning of the photon beam. A special optics with an active mirror has been designed for the TIMEX end-station providing the necessary beam-shaping capabilities for obtaining a well-defined 3–50 micron spot with the desired energy (and fluence) deposited on the sample [14]. Proper diagnostics for the temperature takes advantage of our knowledge on the shape of the pump pulse as described in Refs. [14,16].

The TIMEX end-station [13] is conceived to exploit the unique intensity, energy domain and time structure of the FEL presently under construction, to probe metastable and/or excited matter under extreme conditions. In particular, the energy and intensity of the Fermi@Elettra FEL beam is suitable for an efficient ultrafast heating of most bulk-like dense samples.

As shown in Fig. 1 the end-station design has been maintained very flexible and can accommodate various possible configurations for single-shot experiments including simple far-UV and soft X-ray absorption, and pump and probe experiments using as a probe optical lasers or the FEL pulse (and its harmonics). The optical laser can be also used as a pump. The focussed FEL pulse interacts with the sample installed on a motorized sample manipulator stage working in a Ultra-High-Vacuum (UHV) chamber designed with a variety of windows and feedthroughs (see Fig. 1, upper panel). The 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. Useful diagnostic for initial experiments include a long-distance (LD) microscope for fine micrometric alignments and an infra-red pyrometer [16], but further space is left for additional instrumentation. The main chamber will be equipped with a set of avalanche diode detectors for measurements of direct photon transmission. Fast CCD cameras and diode array detectors will be used for detection of optical ultrashort pulses. The use of an ultrafast streak camera is presently under consideration and tests about the dynamic range and time-resolution of the measurements are also part of the installation program. The sample chamber is fully interfaced through a translation stage with a sample preparation chamber where a fast replacement and surface-science characterization of fresh specimens can be performed.

Both transmission and pump-and-probe experiments using ultrashort photon pulses probing reflectivity and transmission will be possible using proper optical elements (see Fig. 1, lower panel). Suitable windows and space for detectors are left for experiments using harmonics as a probe as well as the possibility of measuring optical absorption of the probe pulses. A table-top supercontinuum probe system [17,18] will be installed for collecting ultrafast optical absorption and reflectivity data in a wide range of wavelengths in a single shot.

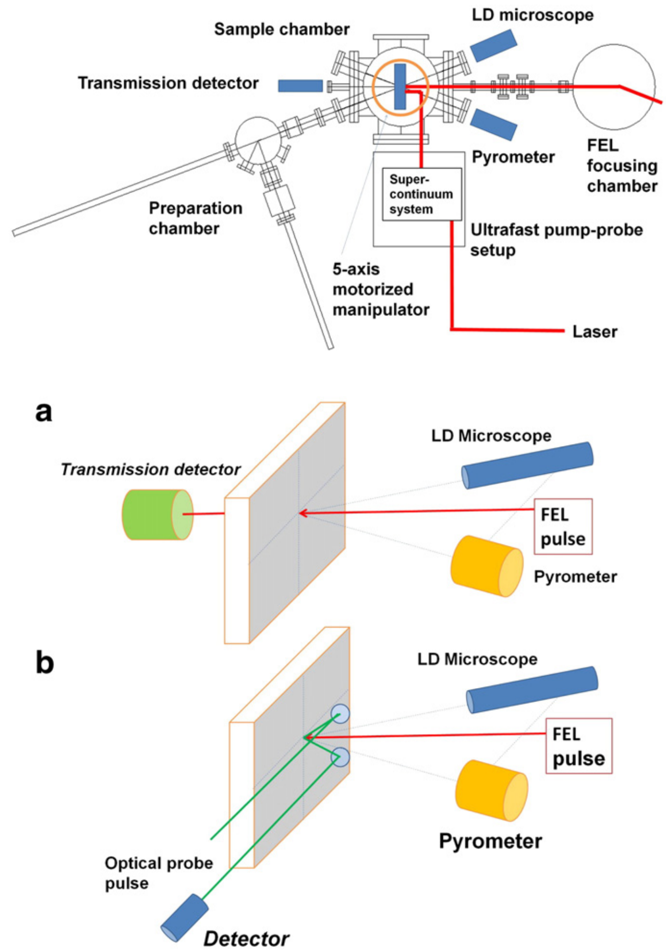

Fig. 1. Upper panel: sketch of the TIMEX end-station under construction at the Fermi@Elettra FEL facility. The FEL pulse is focussed on the sample position through a dedicated mirror set-up [14]. An UHV chamber, connected with a UHV preparation chamber, hosts a 5-axis motorized manipulator allowing for single-shot measurements at 10–100 Hz rate. Diagnostics include a long-distance (LD) microscope for fine micrometric alignments and an infra-red pyrometer [16]. Lower panel: set-up for soft X-ray absorption experiments (a); set-up for ultrafast pump-and-probe optical reflectivity and absorption experiments (b).

3. TIMEX experimental capabilities: pilot pump-and-probe experiments

The end-station design sketched in Fig. 1 can be used for a variety of experiments including simple absorption spectra at different energies (using the FEL energy tunability) and pump-and-probe experiments of optical properties of matter. An important line of research regards pump-and-probe time-resolved studies of the optical and soft X-ray properties of matter providing direct information on surfaces and bulk of samples under extreme conditions. In fact, ultrafast pump and probe experiments in the 0.1–10 ps range, performed under extreme and metastable or non-equilibrium conditions are relevant to a variety of physical and chemical phenomena for which full control and understanding are presently beyond our experimental capabilities. This includes forefront research in high-pressure and high-temperature physics, non-equilibrium and metastable states of matter, applied material studies, understanding of chemical reaction and catalysis paths, planetary interiors, inertial fusion, and various forms of plasma production in which energy is rapidly deposited into a solid.