INTERACTION OF INTENSE ULTRASHORT XUV PULSES WITH DIFFERENT SOLIDS – RESULTS FROM THE TESLA TEST FACILITY FEL PHASE I

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Abstract

Key results of a study of irreversible changes induced at surfaces of metals, semiconductors, and insulators by ultrashort XUV pulses generated at the TTF FEL facility are reported and discussed. Energy spectra of ions ejected from the irradiated surfaces are also reported. Special attention is paid to a difference in the ablation behavior of (semi)conductors and insulators that we have observed. The difference is dramatic while the absorption coefficients are similar for all materials at the TTF1 FEL wavelength.

INTRODUCTION

Before the FELIS [1] experiments were performed at TTF1 FEL in 2001-2002, the shortest wavelengths at which laser ablation had been investigated were 46.9 nm and 125 nm. The 46.9-nm radiation was emitted from a capillary-discharge Ne-like Ar XUV laser [2] and the 125-nm radiation was generated by four-wave-sum-frequency mixing (FWSFM) of a 626-nm dye laser and its second harmonics in Hg vapor [3]. The pulse duration in both cases was in the nanosecond range. A few papers (for a review see [4]) have reported ablation induced at even shorter wavelengths (i.e. in the soft X-ray region) but the radiation used was not coherent. In the present study, not only the laser wavelength is short (< 100 nm) but also the pulse duration is very short (typically 50-100 fs) [5]. The short wavelength and ultrafast coherent beam represent a unique combination. The ablation behavior of a wide variety of materials has been investigated under these irradiation conditions.

In this contribution, key results of a study of irreversible changes induced at surfaces of metals, semiconductors, and insulators by ultrashort XUV pulses generated at the TTF1 FEL facility are reported and discussed. We also report the mass, energy, and spectra of ions ejected from the irradiated surfaces.

Special attention is paid to a difference in the ablation behavior that we have observed for (semi)conductors and insulators. The difference is dramatic while the absorption coefficients are similar for all the materials at the TTF1 FEL wavelength.

Ablation characteristics, i.e. thresholds, etch (ablation) rates, and ablated structure quality, often differ dramatically with conventional UV-Vis-IR lasers, depending on whether the radiation energy is delivered to the material surface in short (typically nanosecond) or ultra-short (typically femtosecond) pulses [6,7]. The FELIS experiment, together with ablation experiments utilizing plasma-based XUV lasers, makes it possible to investigate how ablation characteristics depend on the pulse duration in the XUV spectral region ($\lambda < 100$nm).

EXPERIMENTAL

The samples were irradiated by the SASE-FEL beam emitted by the Tesla Test Facility (TTF) at HASYLAB/DESY in Hamburg [5]. The wavelength was tuned between 85 and 98 nm. Spectral and energy characteristics of the laser beam were measured using photon diagnostics described in details elsewhere [8]. The beam was focused by an elliptical mirror into the interaction chamber as shown in Fig. 1. For more details, see [9].

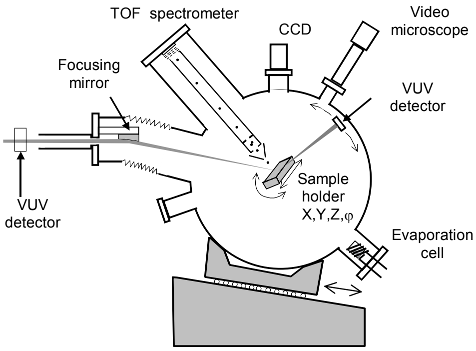

Figure 1: Schematic of the FELIS chamber [9].

Morphological changes at the surface of the exposed samples were investigated by Nomarski, conventional optical, scanning electron, and atomic force (AFM) microscopy. Raman spectra were made with a laser microbeam in the usual backscattering geometry, which enables probing chosen locations on the sample surface. Ion energies and mass spectra were measured using a time-of-flight (TOF) spectrometer equipped with high pass energy filter.

Samples

A number of different samples were irradiated by the focused FEL beam. The bulk samples were: Au, Si, PMMA, Ce:YAG, and SiO_2. We have also irradiated Si crystals polished to optical quality and covered with thin films: 15-nm Au (Au-15), 10-nm and 40-nm amorphous carbon (a-C-10 and a-C-40).

RESULTS

Surface damage

Modification of silicon surfaces exposed to FEL pulses of increasing fluences and recorded by Nomarski microscope is presented in Fig. 2. Each spot was irradiated with 11 shots. The first changes seen are due to modification of the refractive index. An imprinted interference pattern,

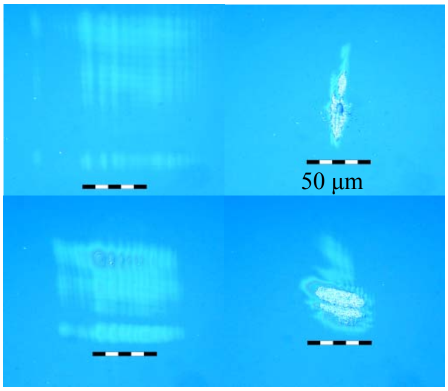

Figure 2: Modification of monocrystalline-Si surfaces imaged by Nomarski microscopy. The fluence increases counterclockwise and is equal to 0.015, 0.04, 0.3, and 1.8 J/cm^2 , respectively.

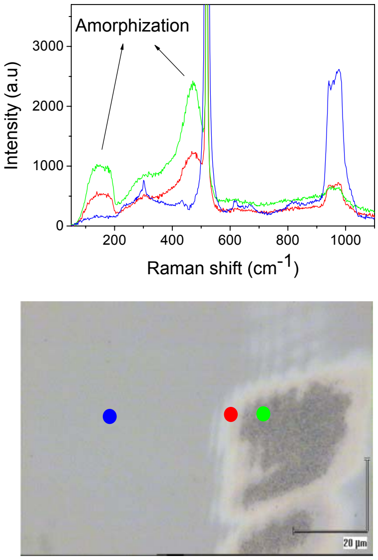

Figure 3: Raman spectra measured at different places on an FEL-irradiated sample of monocrystalline Si.

which is due to diagnostics that were installed in front of the focusing mirror and to slope errors of the mirror surface, is clearly seen. The AFM and TOF measurements do not indicate any morphological changes or ion emission, respectively. Raman spectra however show signs of an amorphous phase in the modified region. The Raman spectra measured at three different locations of the excitation-micro-beam spot are shown in Fig. 3. The amorphous fraction increases towards the center of the crater. The first observable changes were recorded at a fluence of 0.005 J/cm^2.

In Table I we present two different fluence thresholds at which we start to record modifications of the irradiated surface. The first (ΦI) is the fluence at which we notice a change of the refractive index and the second (ΦII) is the fluence at which deformation of the surface is indicated by the AFM. It can be seen that, except for Au-10 and PMMA, the ΦII values oscillate about 0.03 J/cm^2. This is of the order of the fluence, F_c, at which the absorbed energy per atom is equal to the cohesive energy, E_c,

$$F_c = E_c n / \alpha,$$

Table 1: TTF1-FEL surface modification thresholds

Material	ΦI [J/cm^2]	ΦII [J/cm^2]
Au	0.02	0.02
Au-15	0.01	0.01
a-C-40	0.01	0.03
Si	0.005	0.04
SiO_2	0.03	0.03
Ce:YAG	0.02	0.02
PMMA	0.01	0.01

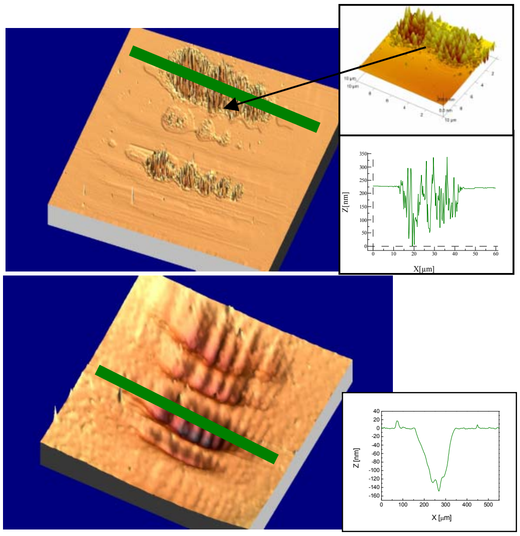

Figure 4: AFM measurements of the silicon (upper) and quartz (lower) samples. Irradiation conditions were similar (fluence ~0.04 J/cm^2). The insets are the crater profiles along the green lines.

where n is atomic density and α is the absorption coefficient. For typical values $E_c \sim 3$ eV/at, $n \sim 5 \times 10^{22}$ at/cm^3, and $\alpha \sim 10^6$ cm^{-1}, $F_c \approx 0.025$ J/cm^2, which corresponds to the values shown in Table 1.

Difference in ablation behavior between (semi)conductors and insulators

Results of the AFM investigations carried out for the crystalline Si and SiO_2 samples are presented in Fig 4. The samples were irradiated with 11 shots at an average fluence of 0.04 J/cm^2. One can notice a dramatic difference in the morphology of both craters. The silicon crater is covered by columnar structures. The peaks of the columns are located above the sample's surface. In the case of crystalline SiO_2 (quartz), the interior of the crater is very smooth and the interference pattern is clearly imprinted into the sample's surface. Exposure of all investigated insulator samples to multiple shots leads to very similar surface pattern when the distance between the sample and the focal spot is the same. Both the morphology of the irradiated surface and the crater depth do not depended on the FEL beam intensity. These features indicate that intense ultra-short pulses of XUV radiation can be used for nano- and micro-machining of insulators.

The ion spectra measured for the Si and SiO_2 samples are presented in Fig. 5. Multiple charged ions were recorded for high-intensity irradiation of Si. The kinetic energy of the ions increases with charge state and reaches the keV range for highly charged ions. Again, there is a clear difference between Si and SiO_2. The high charge states and energetic ions shown for Si are typical of all (semi)conductors. Only singly charged, low-energy ions (~50 eV) were detected for the insulator SiO_2 under all irradiation conditions. Other insulators investigated

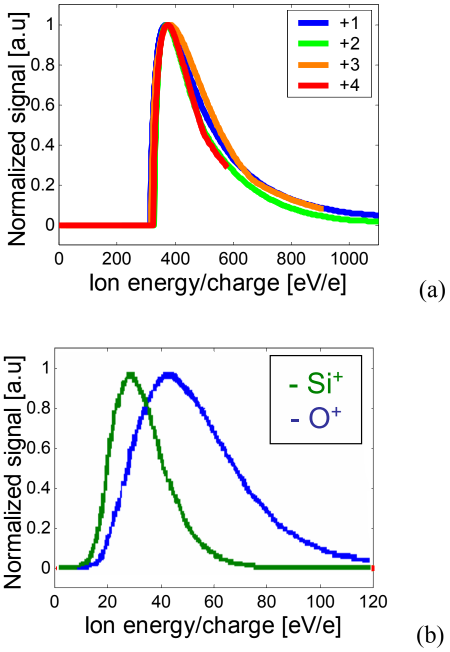

Figure 5: The energy spectra of ions taken with the TOF spectrometer for the silicon (a) and quartz (b) samples. Signals have been normalized. The energy scale has been divided by the ions charge state.

exhibited similar behavior. An interesting feature of the ion spectra observed for all (semi)conductors is that the measured values can be normalized to one curve if the energy of the ion is divided by its charge. This suggests a field emission mechanism.

It is very interesting to notice that the difference in ablation behavior between (semi)conductors and insulators is very pronounced although the absorption coefficients are similar for all the materials at the TTF1 FEL wavelength.

FEL-induced periodic surface structures

Although laser-induced periodic surface structures (LIPSS) were discovered in the middle of the 1960s [10], they remain a subject of extensive research (for a review see [11,12]). According to the nature of their origin, two kinds of LIPSS can be distinguished on laser-irradiated surfaces. In the initial stage of research, most LIPSS observed were ripples, with a spatial period related to the laser wavelength. The ripples, which are called LIPSS of the first kind (LIPSS-I), are created by the interference of the incident laser beam with a wave diffracted by periodic features in the materials' surface. The spatial periods of LIPSS of the second kind (LIPSS-II), however, depend more on laser intensity than on wavelength and are significantly longer than the laser wavelength when a short-wavelength laser is used for irradiation. On the material surfaces irradiated by the focused TTF1-FEL beam, LIPSS-I have been found at the rim of a few of the craters created in a-C and PMMA by 98-nm and 86-nm radiation

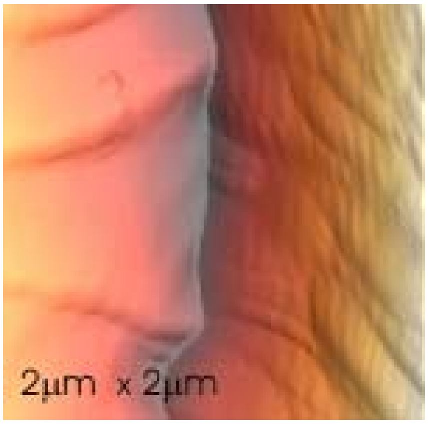

Figure 6: AFM image (tapping mode) of LIPSS at the rim of a crater ablated in PMMA by the tightly focused beam of the TTF1 FEL ($\lambda = 86$ nm, $\phi = 0.5$ J/cm^2; 11 accumulated pulses, 55^o incidence)

respectively [1,13,14]. In Fig. 6, ripples of 70-nm periodicity can be seen on the right side. Currently, 86-nm FEL radiation represents the shortest-wavelength radiation used for production of well-developed LIPSS-I. According to our best knowledge, the shortest wavelength at which LIPSS-I had previously been produced was that of the157 nm F_2 excimer laser [12].

LIPSS-I have not yet been observed on the surfaces of Si, Ce:YAG, or SiO_2 (quartz) irradiated by TTF1 FEL. How might this be explained? Although a possible explanation was discussed in [14], the problem remains unsolved. Contrary to the scarcity of LIPSS-I, LIPSS-II have been found on almost all FEL-irradiated surfaces. LIPSS-II can be attributed to capillary (surface tension) and surface acoustic waves, fixed in laser-melted near-surface material by its re-solidification, and also to Rayleigh-Taylor and Helmholtz-Kelvin instabilities, relaxation of stress, and similar phenomena. All of these processes are expected to be very common at FEL irradiated surfaces, so that the observed high abundance of LIPSS-II is not surprising.

SUMMARY

Results of the study of irreversible changes induced at surfaces of metals, semiconductors, and insulators by ultrashort XUV pulses generated at the TTF1 FEL facility have been reported and discussed. A distinct difference between the behaviors of (semi)conducting materials and insulators was observed. In the case of inorganic insulators, the morphology of the irradiated surface and the crater depth hardly depended on the beam intensity. In contrast, the irradiated silicon surface becomes very rough when the intensity exceeds the damage threshold. There is also a clear difference between insulators and (semi)conducting materials with respect to the spectra of ejected ions. Highly-charged, energetic ions (~ a few keV) were typical for (semi)conductors. Only singly-charged, low-energy ions (~50 eV) were detected for insulators under all irradiation conditions. LIPSS-I have been found at the rim of a few craters created in a-C and PMMA by 98-nm and 86-nm radiation, respectively. LIPSS-II have also been found. Their spatial periods depend more on laser intensity than wavelength and are significantly longer than the laser wavelength, for short-wavelength lasers.

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