

Ablation of solids using the femtosecond XUV free electron laser FLASH

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Ultrafast pulsed excitation of solids provides a unique way of depositing energy into materials and to create states of strong electronic excitation and high temperature and pressure. With the initial deposition of energy a complex chain elementary physical processes is triggered which can lead to structural changes and material removal on very rapid time-scales, and often along unusual, non-equilibrium pathways. Besides their importance from a fundamental physics point of view these processes are also of major relevance in many technological applications like short-pulse laser-processing. Up to now mainly femtosecond optical lasers have been used for material excitation, but the interpretation of experimental data is often difficult because of the highly non-linear nature of the deposition of optical energy in the intensity range of interest ($> 10^{12}$ W/cm²).

New possibilities for both generating and probing high energy density states of matter are emerging with the recent advent of short-wavelength LINAC-based light sources. Among these future light sources the XUV free electron laser FLASH is worldwide the first SASE-FEL operating in the 6 - 100 nm wavelength range. The irradiation of solid materials with such short-wavelength femtosecond pulses offers a number of advantages. First of all it permits a high degree of electronic excitation but with a strongly reduced influence of optical non-linearities (i.e. multi-photon absorption, free carrier absorption). Moreover, for frequencies higher than the plasma frequency but lower than the frequency of the inner-shell absorption edge, the absorption depth for some materials can be rather long. Therefore, ultrashort XUV-pulses allow the preparation of well-defined excitation conditions in relatively large sample volumes as compared to femtosecond optical pulses.

Here we report on the results of first experiments performed at FLASH on the interaction of ultrashort high intensity ($10^{12} - 10^{14}$ W/cm²) XUV-pulses with solid surfaces. In an XUV-pump/optical probe experiment picosecond optical imaging has been used to follow the dynamics of short-pulse XUV-induced phase transitions and ablation. Experiments were performed at beam line BL2 of the FLASH facility with the FEL operating at a wavelength of 32.5 nm and a pulse duration of approximately 25 fs (FWHM).

A typical set of data is displayed in Fig. 1 which shows a sequence of time-resolved micrographs from a Si surface irradiated at a fluence of 2.5 J/cm². For very short delay times a pronounced increase of the reflectivity to more than twice the initial value of unexcited Si is observed which can be attributed to a rapid melting transition of the material. Due to the high fluence material removal (ablation) becomes visible as early as 10 ps, as evidenced by the decrease of the reflectivity in the center of the spot, where the excitation is strongest. For times of up to approximately one nanosecond, the size of the low reflectivity region increases while the size of the

molten area remains unchanged. On a nanosecond time-scale resolidification of the material sets in at the periphery of the spot where the excitation fluence is low. On the same time-scale an additional, circular feature appears in the center of the spot which indicates melt expulsion induced by the recoil pressure of the ablating material (so-called *piston-effect*). Even after 18 ns ablation has not come to an end, and the irradiated surface has not reached its final state as can be seen from the last frame of the series taken a few seconds after the irradiation.

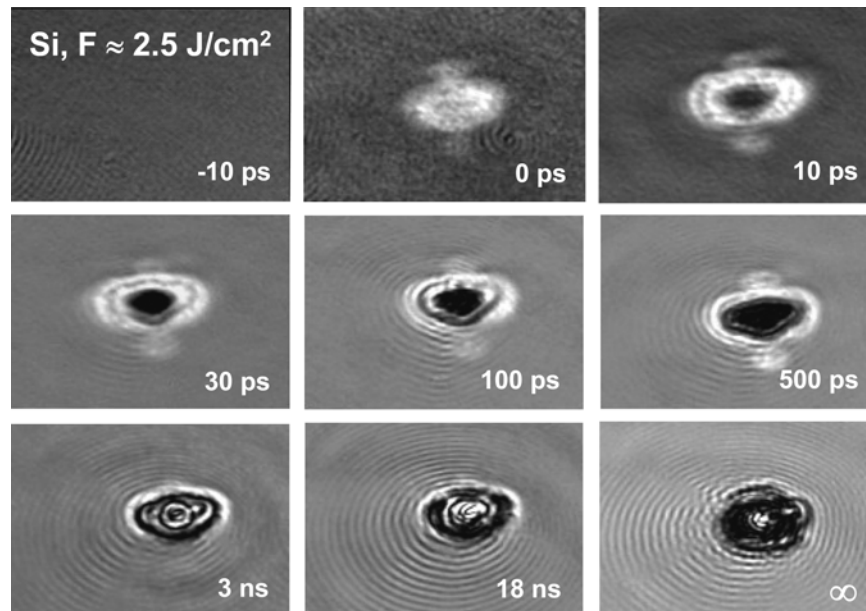


Figure 1: Time-resolved micrographs of a Silicon surface irradiated with a <50 fs VUV-FEL pulse ($\lambda = 32$ nm) and a fluence of 2.5 J/cm^2 ; frame size is $120 \times 80 \mu\text{m}^2$.

The time-resolved measurements were supplemented by a characterization of the permanent structural modifications of the irradiated surfaces. A comparison with femtosecond optical excitation reveals distinct differences in the material response which we attribute to the absence of absorption non-linearities and the increased penetration of the XUV-light in the materials (Si and GaAs) under study [1]. Future experiments with improved temporal resolution will allow us to address directly the specific aspects of electronic relaxation after excitation with high energy photons and to investigate the dynamics of the resulting rapid phase transformations.

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References

- [1] N. Stojanovic, D. von der Linde, K. Sokolowski-Tinten, U. Zastrau, F. Perner, E. Förster, R. Sobierajski, R. Nietubyc, M. Jurek, D. Klinger, J. Pelka, J. Krzywinski, L. Juha, J. Cihelka, A. Velyhan, S. Koptyaev, V. Hajkova, J. Chalupsky, J. Kuba, T. Tschentscher, S. Toleikis, S. Düsterer, and H. Redlin, Appl. Phys. Lett. (in print, 2006).