Modeling of soft X-ray induced ablation in solids

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ABSTRACT

Powerful free electron lasers (FELs) operating in the soft X-ray regime are offering new possibilities for creating and probing materials under extreme conditions. We describe here simulations to model the interaction of a focused FEL pulse with metallic solids (niobium, vanadium, and their deuterides) at 13.5 nm wavelength (92 eV) with peak intensities between $10^{15}$ to $10^{18}$ W/cm\textsuperscript{2} and a fixed pulse length of 15 femtoseconds (full width at half maximum). The interaction of the pulse with the metallic solids was modeled with a non-local thermodynamic equilibrium code that included radiation transfer. The calculations also made use of a self-similar isothermal fluid model for plasma expansion into vacuum. We find that the time-evolution of the simulated critical charge density in the sample results in a critical depth that approaches the observed crater depths in an earlier experiment performed at the FLASH free electron laser in Hamburg. The results show saturation in the ablation process at intensities exceeding $10^{16}$ W/cm\textsuperscript{2}. Furthermore, protons and deuterons with kinetic energies of several keV have been measured, and these concur with predictions from the plasma expansion model. The results indicate that the temperature of the plasma reached almost 5 million K after the pulse has passed.

**Keywords:** X-ray free electron laser, plasma, ion acceleration, ablation, non–local thermodynamics equilibrium

1. INTRODUCTION

Any sample placed into a focused beam of an X-ray free-electron laser will eventually turn into plasma at some point. Damage to the sample is driven by direct photoionization followed by various de-excitation processes, including the release of Auger and shake electrons. Electrons ejected from atoms will produce extensive secondary electron cascades in condensed materials. Most cascade processes come to completion within 1-10 femtoseconds, following the primary ionisation event. The temperature of the electron gas is very high at this point. The coupling between the electron gas and the ions in the sample leads to a transfer of energy to the ions and an equilibration of the electron and ion temperatures over a period of a few hundred femtoseconds. The resulting plasma temperature is lower than the initial electron temperature but it can still reach millions of K. Expansion of the sample during this period is driven by the increased kinetic energy of the ions and by electron pressure. Under these conditions, vaporization of micron-sized objects can take a few picoseconds,\textsuperscript{1} and ablation of bulk solids requires only a few nanoseconds.\textsuperscript{2} Re-solidification of surface layers in ablated solids takes an additional 100 ns.\textsuperscript{3}

We present here a theoretical study on previously performed experiments at the FLASH free electron laser\textsuperscript{4, 5} on metallic solids.\textsuperscript{6} The experiments used a sub-micron focusing mirror\textsuperscript{7, 8} to reach intensities in excess of $10^{17}$ W/cm\textsuperscript{2} on the surface of the samples, including niobium (Nb), vanadium (V) and their deuterides (NbD, VD). We measured ion time-of-flight (TOF) spectra directly off the sample surface to determine the energy of ions ejected from the sample, and characterised the resulting ablation craters by optical microscopy, electron microscopy (EM), and atomic force microscopy (AFM). We find a correlation between ion acceleration driven by

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plasma expansion and processes that determine the depth of the observed craters by linking both to a saturation of the average ionization on the plasma surface with increasing intensity of the pulse. This results in saturation of proton energies at high intensities and enables the deposition of energy deep into the sample through a transient transparency in the surface.

2. COMPUTER MODELING

The experimental results and measurements\(^6\) need explanation through theory. Our theoretical approach contains extensive modeling of the interaction between ultra-short and ultra-intense soft X-ray laser pulses from FLASH and metallic samples (Nb, V, NbD and VD). The simulations have been performed using Cretin\(^9,10\), a multidimensional non-local thermodynamic equilibrium (non-LTE) plasma code, which is based on an established plasma model for low-density hot matter\(^11,12\). The code calculates atomic kinetics and populations affected by the laser-matter interaction and complements these with calculations on radiation and heat transfer to provide detailed information on relevant changes in a sample during and after exposure to a laser pulse. Modeling of the rapid changes in electron and ion temperatures in the sample using non-LTE dynamics has been performed previously on other samples, on solids (B\(_4\)C, SiC, and Si) and soft biological matter (cells)\(^13,14\). Cretin uses the so called screened hydrogenic model, which assumes that the wave function of each electron can be calculated as a hydrogen-like atom, with screening constants that mimic the effect of the other electrons\(^15,16\). This method has been shown to give an accuracy within 25\% of the values calculated with the Hartree-Fock-Slater theory. The model explicitly follows all electronic transitions between different bound states, as well as bound-free transitions. The rates for these processes are calculated and a self-consistent rate equation is used to determine the evolution of the atomic populations. Lowering of the ionization potential is modeled through the Stewart-Pyatt formula\(^17\), a commonly used approximate model which has been evaluated against more detailed models as well as against experiments\(^18\). The plasma front expansion into vacuum is then calculated using a self-similar isothermal fluid model\(^19\). This model gives a good description of the acceleration of protons off a surface in optical laser-driven experiments, over a wide range of parameters\(^20\), and is extended in our case into the soft X-ray regime.

Figure 1 shows the setup for the computer simulations with Cretin. A sample material of variable thickness
Figure 2. Simulations of heating and absorption dynamics in VD performed with Cretin in one dimension. The simulated laser pulse has a photon energy of $E_{ph} = 92 \text{ eV}$ (corresponding wavelength $\lambda = 13.5 \text{ nm}$) with an intensity of $10^{17} \text{ W/cm}^2$. Shown in (a) is the average ionization per atom ($\bar{z}$) and in (b) the electron temperature ($T_e$) as a function of time and depth in the sample.

(1–10 $\mu$m) is modeled in one dimension and is segmented typically into 250 zones of equal size. These zones are interlinked through radiation and heat transfer and contain niobium, vanadium, or their deuterides at solid densities. Heat transfer is calculated based on a two-temperature model that follows the coupled temperature evolution of the ions and electrons. For the short FEL laser pulse a Gaussian temporal profile with 15 fs FWHM is assumed. Simulations were performed with a photon energy of $E_{ph} = 92 \text{ eV}$ (corresponding wavelength of 13.5 nm) and with varying intensities ranging from $10^{15}$ to $10^{18} \text{ W/cm}^2$, covering the experimental conditions of the FLASH experiment. The simulated cold absorption spectra for Nb and V at the given wavelength were matched with the theoretical values from Henke et al. The time evolution in the simulation is done in steps of 0.01 fs, with a total simulation time up to 1 ps.

Figure 3. Temporal development of the simulated electron ($T_e$) and ion temperatures ($T_i$) in VD after irradiation with a 15 fs (FWHM) soft X-ray pulse with $\lambda = 13.5 \text{ nm}$ and an intensity of $10^{17} \text{ W/cm}^2$. $T_e$ increases with the onset of the pulse and reaches a maximum directly after the pulse, whereas $T_i$ evolves slower and approaches a maximum value long after the pulse has passed. The timescale for the equilibration of the electron and ion temperatures is $\sim 100$ fs.

The dynamics of the laser-plasma interaction during one simulation on VD can be seen in Figure 2 for an intensity of $10^{17} \text{ W/cm}^2$. The evolution of the average ionization per atom ($\bar{z}$) and electron temperature ($T_e$) can be followed throughout different time scales. During illumination with the 15 fs long X-ray pulse, photoionization, Auger emission, shake processes and secondary impact ionisations dominate in the sample. Thermal equilibration occurs on intermediate time scales, 10–100 fs, when the electron and ion temperatures equilibrate.
Figure 4. Average ionization per atom ($\bar{z}$) and electron temperature ($T_e$) for VD as a function of depth simulated with CRETIN. Simulations were performed with a 15 fs long soft X-ray pulse of intensity $10^{17}$ W/cm$^2$ ($\lambda$=13.5 nm). The modeling allows for an individual examination of the effects of radiation transfer (RT), electron thermal conduction (TC) and hydrodynamic expansion (HE) on $\bar{z}$ and $T_e$ for different timescales. a) and b) Evolution of $\bar{z}$ and $T_e$ directly after the pulse. Radiation transfer and electron thermal conduction dominate in the sample over a short period of time, and at this stage the material is affected only to a shallow depth. c) and d) 500 fs after the pulse, material transport through additional contributions from hydrodynamic expansion have to be taken into account affecting deeper layers of the sample.

The equilibration can also be seen in more detail in Figure 3. Radiation and heat transport dominate on longer time scales, 100 fs – 1 ps, and start ionizing atoms and heat the sample in the bulk.

We examine different interaction processes during and after the pulse (Figure 4). Energy exchange between zones due to radiation transfer (RT) or electron thermal conduction (TC) can be assessed individually to describe the heating of the sample. As can be seen in Figure 4(a) and (b) for $10^{17}$ W/cm$^2$, the heating in VD is promoted by radiation transfer in the first few femtoseconds after the pulse. Radiation from the initial laser pulse and reemission in the sample contribute significantly to the depth of energy deposition at short timescales. Competing with this is the rapid equilibration of the electron temperature $T_e$ in the material due to thermal conduction which results in a temperature drop in the surface region. The initial heating processes act on a faster timescale than the actual plasma expansion. Long after the pulse, when material transport becomes dominant, the effects of hydrodynamic expansion (HE) have to be taken into account (Figure 4(c) and (d)). On long timescales (> 500 fs), hydrodynamic expansion may contribute further to the deformation of the sample and ablation into larger depths that are not reached when only taking the initial heating processes into account.

3. DISCUSSION

Our simulations showed that the temperature at the surface of the solids reached values between 100 – 400 eV for intensities above $10^{16}$ W/cm$^2$ (Figure 5). After electron-ion equilibration on time scales of about 100 fs, all ions in the simulation reached similar high temperatures. The ions at the plasma front were accelerated towards
higher energies exceeding the electron temperatures. Over the range of intensities studied in the experiment, the measured energies of the hydrogen from the acceleration reached values between 1-5 keV (Figure 5(b)). This is consistent with the calculated maximum ion front energy in an expanding plasma. These calculations of proton acceleration are based on the self-similar isothermal fluid model for plasma expansion into vacuum. It also describes the highly charged Nb and V ions, which reach up to 20 keV. At intensities exceeding $4 \times 10^{17}$ W/cm$^2$, saturation in the simulated average ionization $\bar{z}$ at the surface of the plasma is visible in Figure 6(a). This hints at a transient transparency at the sample surface with increasing intensity of the laser pulse. Furthermore, electron temperatures above 500 eV enable ionization of the vanadium L-edge. Below $4 \times 10^{17}$ W/cm$^2$, deeper electronic levels are accessed through collisional ionization, which on a time scale of 100 fs after the pulse leads to a decrease in electron temperature and an increase in average ionization. At higher intensities, ionization saturates while the electron temperature increases (Figure 6(b)). On times scales of 100 fs, the electron-ion thermalization will reduce the electron temperature.

To model the ablation process and the observed crater depths in the experiment, we introduce the concept of critical depth. We note that the attenuation length for 92 eV photons in V and Nb is different by a factor of 5, when calculated based on the tabulated cold absorption coefficients (Figure 7). This is in contrast with the observed crater depths, which at high intensities are similar for all the samples containing V and Nb. Following our previous model, we define the critical charge density as the point where the density of free carriers, electrons is high enough to destabilize the lattice. This happens at $N \approx 10^{22}$ cm$^{-3}$ and considering the atomic ratios and crystal densities of our samples, this corresponds to an average ionization state per atom of $\bar{z} = 0.16$ for VD and $\bar{z} = 0.20$ for NbD. The simulated time evolution of the critical charge density is presented in Figure 8. After 1 ps the simulated critical depth approaches the measured crater depths for intensities around $10^{16}$ W/cm$^2$. At the same time, this critical density does a less satisfactory job at describing the crater depths towards higher intensities $10^{17}$ W/cm$^2$.

As an alternative to the concept of critical depth, we used the experimental data on the crater depth to determine the critical temperature for the equation-of-state in a two-phase vaporization model, similar to previous studies by Hau-Riege et al. Starting with the experimentally measured crater depth for various samples, we determine the electron temperature at that certain depth. We find that the electron temperature from the simulations after the X-ray pulse ends, varies between 0.3 and 0.6 eV for all the samples. These temperatures are in the same range as other critical temperatures in solids, which have been obtained at lower intensities. The large variation in the electron temperature values comes from the large spread in the crater depths obtained experimentally at a fixed intensity.
Figure 6. Simulated average ionization and temperature of the plasma at the surface of the sample for intensities exceeding $10^{16}$ W/cm$^2$. (a) Average ionization per atom for VD and NbD as a function of intensity, shown at different times during the simulations with CRETIN. After 100 fs, an increase in ionization is visible and can be traced to collisional ionizations in the plasma. (b) Intensity dependent evolution of the electron temperature in VD at different times throughout the laser-matter interaction. The ionization levels for V have been added (black dashed lines). Starting at $4 \times 10^{17}$ W/cm$^2$ deeper electronic levels can be accessed through ionization processes in the hot plasma. The vanadium L-edge is reached with $T_e$ exceeding 500 eV.
Figure 7. Calculated photon transmission into the cold sample for V and Nb based on their cold absorption coefficients for 92 eV photons, considering an initial pulse with $10^{12}$ photons. V has a 5 times higher cold absorption than Nb resulting in a smaller corresponding attenuation length of only 40 nm (200 nm for Nb).\textsuperscript{21} The experimentally observed crater depths and diameters are similar for all V and Nb samples at high pulse intensities,\textsuperscript{6} showing that cold absorption is not suitable for describing the observed craters.

Figure 8. Critical depth as a function of pulse intensity for different materials (V, VD, Nb and NbD) simulated with Cretin. The X-ray pulse is modeled to have 15 fs FWHM and $\lambda=13.5$ nm. Shown are the simulated depths where the critical density of electrons N=$10^{22}$ cm$^{-3}$ is reached after 0.1 ps and 1.0 ps. Furthermore, the depths where the critical electron temperature approaches $T_e=0.3$ eV and 0.6 eV after 0.1 ps are also presented. NbD shows a different behavior than VD at high intensities. This is likely due to deviations in absorption lengths for the different materials\textsuperscript{21} and a different development of transient transparency in the samples.\textsuperscript{6}

When comparing the experimental results of crater depths with the critical depths obtained from the simulations, there is an apparent discrepancy towards high intensities. The 1D simulations show that the critical depth, whether defined based on the average ionization or the electron temperature, increases dramatically with higher pulse intensities. However, the experimental results hint at a saturation in crater depth with high intensities, which becomes more clear when high intensity is achieved through tighter focus. This suggests that our 1D description is no longer adequate when the focus of the laser is comparable or smaller than the critical depths. A proper description of transport phenomena in the bulk, which could explain both the crater depths and diameters, requires simulations at higher dimensions and possibly an introduction of high field effects.

4. SUMMARY

Our theoretical work describes the damage and ion acceleration processes that govern the interactions between a tightly focused soft X-ray laser beam and solid metal and deuterated metal targets. Samples irradiated with individual pulses from a soft X-ray FEL focused down to sub-micrometer spot size will turn into a hot plasma...
with temperatures of about 400 eV when intensities exceeding $10^{17}$ W/cm$^2$ are reached. This creates conditions where the initial heating is sufficiently high to promote ablation several μm into the sample. The craters are formed after plasma expansion into vacuum, a process during which light ions are accelerated to about 5 keV. Metal ions reach multiple charge states and energies of about 20 keV. These results are in agreement with the measurements at FLASH. The onset of saturation in ion acceleration as well as crater formation is observed with increasing intensities and the effects become more pronounced exceeding $10^{17}$ W/cm$^2$. At the same intensities our simulations display the onset of saturation effects in the electron temperature and average ionization per atom. The occurrence of a transient surface transparency during the pulse would enable the incoming X-rays to penetrate deeper into the sample and deposit their energy into a larger volume. This is one possible explanation for the observed saturation effects. Discrepancies between experimental results of crater depths and simulated values of critical depths at high intensities suggest that a 1D approach might not be entirely adequate to describe the transport phenomena in the bulk. Possible high field effects at such high intensities have to be evaluated to account for the observed saturation in ion energies and crater depths.

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