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Time-resolved pump-probe analysis of metal ablation using single and double ultrashort laser pulses

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Abstract

In recent years several works have been investigating the temporal distribution of pulse energy with the aim of maximizing ablation efficiency in terms of energy specific ablation volume by applying double pulses or pulse bursts. Here we study the energy specific ablation volume of double pulses in dependency of increasing pulse separation on aluminum samples. To avoid multi-pulse incubation effects the double pulse irradiation is repeated only three times at one position. Ultrafast pump-probe ellipsometry and microscopy are presented in order to study the surface material motion after single laser pulse impact. The comparison of time-resolved measurements and double pulse ablation volumes suggests that between 5 ps and 20 ps an interaction of the second pulse with the rarefaction wave decreases double pulse energy specific ablation volume. For longer double pulse spacing up to 1 ns the decrease can be attributed to re-deposition of ablated material.

Keywords: ultrashort pulses, double pulses, ellipsometry, pump-probe, ablation, metals

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1. Introduction

Ultrashort laser pulses in the ps and fs regime are used in various material processing applications such as, selective thin film removal [1] and metal micro processing [2].

Experiments performed on copper samples with double pulses, showed that for temporal pulse spacing t_P below 1 ps, the ablation depth of the double pulses remains maximal. However, when t_P exceeds 1 ps, the double pulse ablation depth drops rapidly and reaches the ablation depth of a single pulse at t_P≈10 ps. Increasing t_P further to values ranging between 10 ps and 100 ps, results in the double pulse ablation depth dropping below the value of the single pulse ablation depth [3]. In 2018, Förster et al. performed similar double pulse experiments on copper samples for t_P ranging from 100 fs to 10 ns. For temporal pulse spacing up to 100 ps, their results agree qualitatively with the results obtained in reference [3]. In their experiments, the observation of the double pulse ablation depth dropping below that of a single pulse continues to temporal pulse spacing of t_P≈2 ns. A further increase of t_P results in an increase of the double pulse ablation depth, eventually approaching the single pulse level at t_P≈10 ns. The state of the art presented above clearly shows that double pulse laser processing is highly inefficient for t_P ranging between 10 ps and 10 ns.

Attempts have been made to explain the decrease of the double pulse ablation depth with increasing t_P. Povarnitsyn et al. performed hydrodynamic modeling of double pulse experiments, showing that the double pulse acts as a single pulse for $t_P \ll t_{ei}$, where t_{ei} denotes the electron-ion relaxation time. When t_P is on the order of t_{ei}, the second pulse suppresses the rarefaction wave induced by the first pulse, resulting in a decreased ablation depth. For $t_P \gg t_{ei}$ it is argued that only the first pulse contributes to the formation of the ablation crater, while the second pulse is absorbed by the ejected ablation plume [4]. While the above findings from hydrodynamic modeling can certainly explain the drop in double pulse ablation depth in the first few ps, they can't explain the double pulse ablation depth dropping below that of a single pulse. However, previously reported experiments and simulations suggest that the drop below the single pulse level can be explained by re-deposition of the ablation plume [5, 6].

In this study, double pulse experiments with temporal pulse spacing ranging between 0 ps and 1000 ps are performed on aluminum samples in order to identify the critical temporal double pulse spacing regimes. To avoid multi-pulse incubation effects as far as possible the double pulse irradiation is repeated only three times at one position. In addition, ultrafast pump-probe ellipsometry and microscopy are performed to compare the surface material motion with pulse separation dependence of the double pulses energy specific ablation volume (ESAV).

2. Materials and Methods

All experiments were performed using a linear polarized Nd:Glass laser source, which emits 680 fs (FWHM) pulses at a central wavelength of 1056 nm and a repetition rate of 500 Hz. In each setup the pulses were focused on the sample surface using a lens with a focal length of 150 mm, producing a focal radius of 15 µm ($1/e^2$). Single pulses were selected from the pulse train using a fast mechanical shutter.

Double pulses were generated from the frequency doubled single pulses with the aid of a Michelson interferometer, resulting in perpendicular polarized double pulses with temporal pulse spacing ranging between 0 ps and 1 ns. A detailed description of the pump-probe microscopy and ellipsometry setups can be found in [7, 8]. The experiments were performed on aluminum samples, which were polished to achieve a smooth surface with an average surface roughness of a few nm. Ablation Craters have been produced by three successive double pulses, with a temporal spacing of 1 s between the individual double pulses to minimize multi-pulse incubation effects. In order to account for the crater morphology, the ablation volume instead of the ablation depth were measured and divided by the incident laser pulse energy to obtain the ESAV. All ESAV results were averaged over a sample size of three craters.

3. Results and Discussion

3.1. Double Pulse Experiments

The single pulse ablation threshold fluence of the investigated aluminum samples was determined to be F_{thr}=0.72 J/cm^2 using the D^2-Method [9]. Single pulse as well as double pulse experiments with t_P ranging between 0 ps and 1 ns were performed in order to identify the critical pulse separation regimes described in section 1 for aluminum samples. Figure 1 shows the measured double pulse ESAV for t_P ranging between 0 ps and 1 ns.

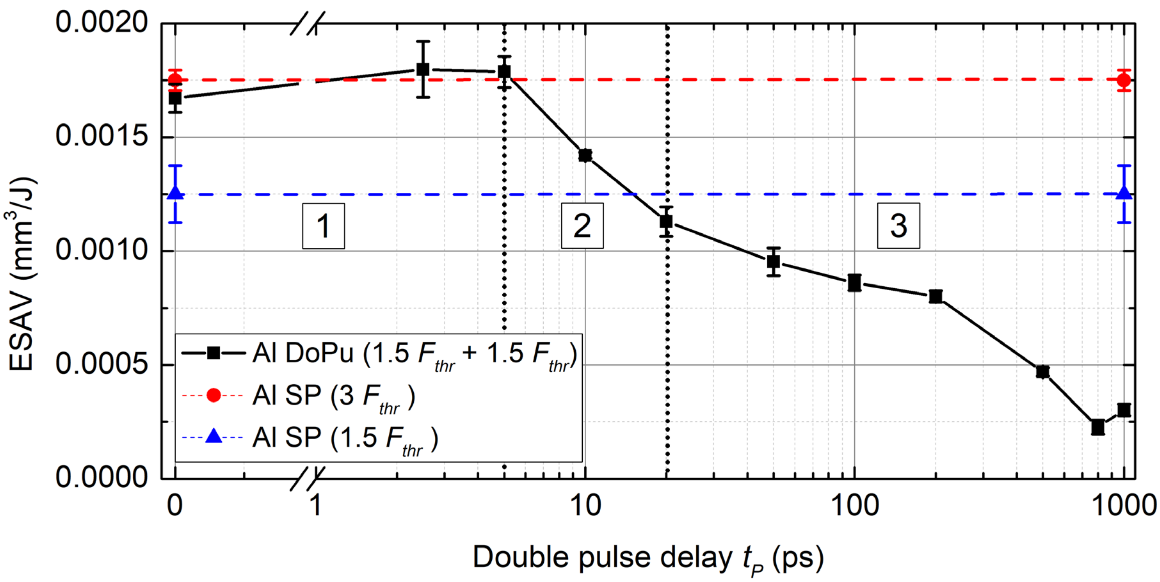

Fig. 1. Double pulse experiments performed on aluminum samples with t_P ranging between 0 ps and 1000 ps. Double pulses with a fluence of F_0=1.5·F_{thr} each are indicated by black squares. The values for single pulse ablation with a fluence of F_0=3·F_{thr} (red circles) and F_0=1.5·F_{thr} (blue triangles) are shown for comparison.

The double pulses (black squares) are composed of two pulses exhibiting a peak fluence of F_0=1.5·F_{thr} each. The ESAV values of single pulses with peak fluences of F_0=3·F_{thr} (red circles) and F_0=1.5·F_{thr} (blue triangles) are also shown for comparison. In general, three temporal regimes can be distinguished: In the first regime, which ranges from t_P=0 ps to t_P=5 ps the double pulse ESAV remains at the value of a single pulse carrying the same energy. Moving on to the second regime, the double ESAV decreases significantly, reaching the value of a single pulse with F_0=1.5·F_{thr} at t_P=20 ps. The third regime, which lasts until the maximum measured t_P value is characterized by a decrease of the double pulse ESAV below the value of a single pulse with half peak fluence. The results depicted in Figure 1 agree qualitatively with the results obtained in earlier multi-pulse works on copper samples [3, 5].

3.2. Pump-Probe Ellipsometry

Figure 3 shows the temporal evolution of the extinction coefficient *k* (imaginary part of the complex refractive index) for delay times ranging between 15 ps before pulse impact to 1 ns after pulse impact for a laser peak fluences F_0=2.25·F_{thr}. A detailed description of how *k* is obtained from the pump-probe ellipsometry measurements can be found in [8].

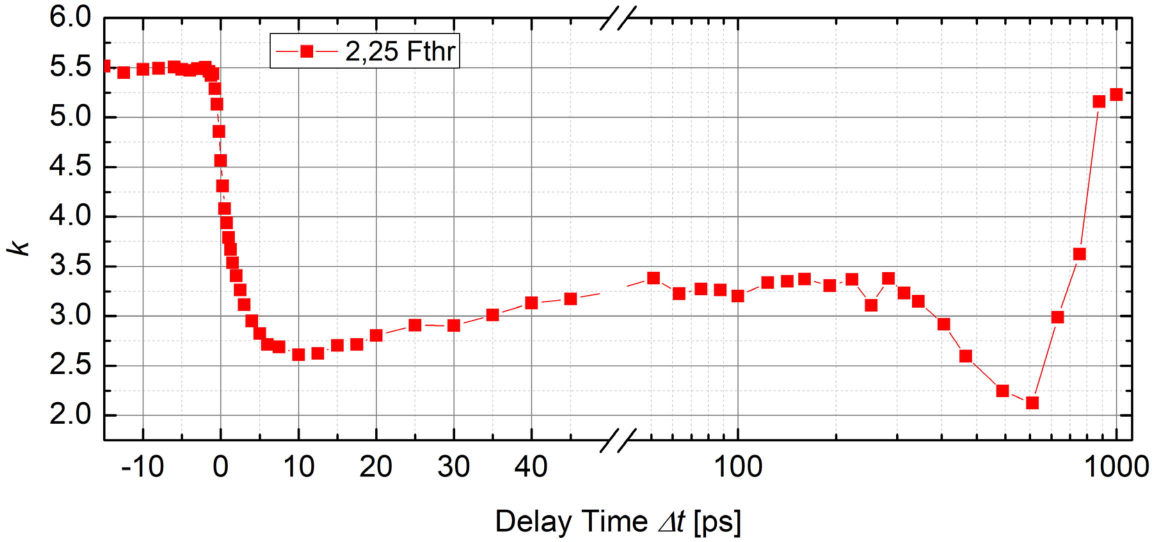

Fig. 2. Transient evolution of the extinction coefficient *k*, obtained from pump-probe ellipsometry experiments performed on aluminum samples with *Δt* ranging between 0 ps and 1000 ps and a laser peak fluences of F_0=2.25·F_{thr}.

A steep decrease of the extinction coefficient *k* from its initial value *k*≈5.5 nm is observed almost instantaneously after laser pulse impact at *Δt≈0* ps. The steep decrease results in a local minimum at *Δt≈5* ps with a value of *k*≈2.5 nm. After the initial decrease, the values of *k* recover before reaching another local minimum with a value of *k*≈2.0 nm at *Δt*≈700 ps. Finally, at delay times of *Δt*≈900 ps, the extinction coefficient approaches its initial value.

3.3. Pump-Probe Microscopy

Experimental data of the relative sample surface reflectivity change *ΔR/R* is shown in figure 4 for a single pulse laser peak fluence of F_0=2.5·F_{thr} and delay times ranging between -15 ps and 1000 ps. The figure shows, that the initial rapid decrease of *ΔR/R* is followed by an oscillatory behavior, which starts at *Δt*≈50 ps and lasts until *Δt*≈600 ps.

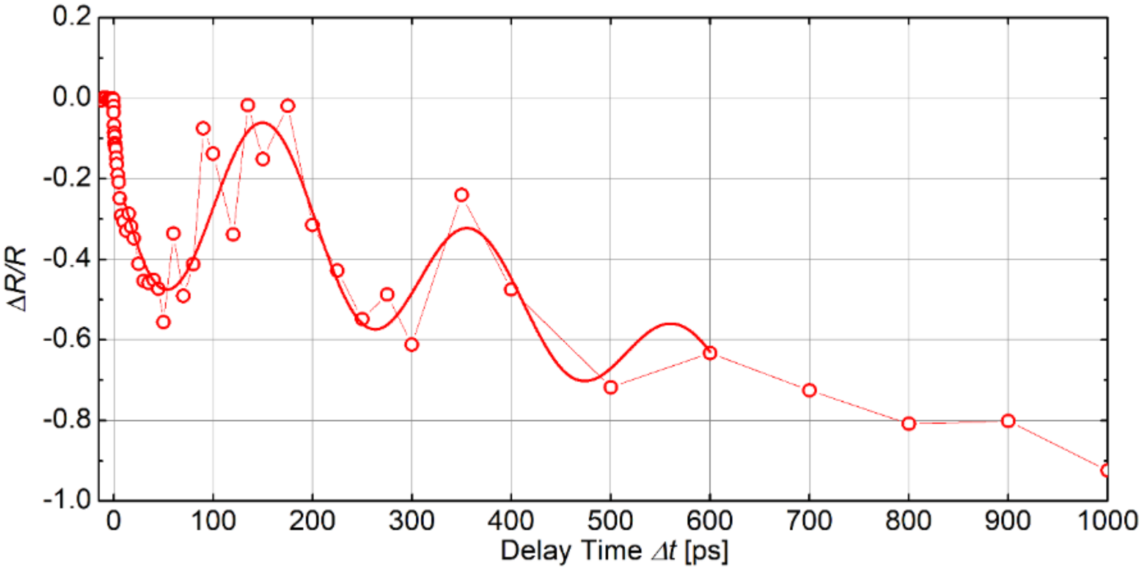

Fig. 3. Transient evolution of the relative reflectivity change, obtained from pump-probe microscopy experiments performed on aluminum samples with delay time *Δt* ranging between -15 ps and 1000 ps at a laser peak fluences of F_0=2.5·F_{thr}.

3.4. Discussion

As depicted in figure 1, the double pulse ESAV stays unchanged in regime 1 for t_P≤5ps. Since the electron-ion relaxation time of aluminum is estimated to be of the order of 3-4 ps [10], the experimental results shown in this paper agree well with the hydrodynamic simulation result that a double pulse acts as a single pulse for a double pulse spacing which is on the order of the electron-ion relaxation time [4]. The pump-probe ellipsometry measurements show a steep decrease of *k* in regime 1 until *Δt≈5* ps. This steep decrease of the extinction coefficient after pulse impact was attributed to a density decrease of the irradiated surface, induced by the build-up of a rarefaction wave [11, 12]. Therefor it can be concluded that the complete build-up of the rarefaction wave takes about 5 ps and that the second pulse is not disturbed by the emerging rarefaction wave.

In regime 2 for 5 ps ≤ t_P ≤ 20 ps, the double pulse ESAV drops to the level of a single pulse carrying half the double pulse fluence. On this time scale, the rarefaction wave induced by the first pulse has been fully established, which can be concluded from *k* being approximately constant in regime 2*.* This result indicate that the second pulse is disturbed by the rarefaction wave induced by the first pulse only after it has been completely established.

Regime 3, which ranges from 20 ps≤t_P≤1 ns, is characterized by the double pulse efficiency dropping below that of a single pulse. The onset of the drop below the single pulse level approximately coincides with the beginning of the oscillatory feature observed in the pump-probe microscopy measurements (see figure 3). This oscillatory behavior of the relative sample surface reflectivity was observed in various works and is attributed to Newton rings, which are directly related to the surface material motion [13]. Here it can be concluded that the second pulse interacts with the moving ablation front in regime 3 and therefor can cause a re-deposition of the ablated material.

4. Conclusion

In conclusion double pulse experiments on aluminum have been carried out, and a special emphasis has been given to avoid incubation effects and to take the crater morphology into account. Three distinct temporal double pulse spacing regimes have been identified for aluminum. For the first regime (0 ps≤t_P≤5 ps) it was shown that a double pulse acts as a single pulse for double pulse spacing below 5 ps and that the build-up of the rarefaction wave doesn't impact the double pulse ESAV. The decrease of the double pulse ESAV in the second regime (5 ps≤t_P≤20 ps) was attributed to the interaction of the second pulse with the completely established rarefaction wave. In the third regime (20 ps≤t_P≤1 ns) the second pulse was found to interact with the moving ablation front, possibly resulting in a re-deposition of ablated material.

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