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Adaptive control of femtosecond laser ablation plasma emission

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ABSTRACT

The influence of temporal pulse shaping on plasma plume generated by ultrafast laser irradiation of aluminum is investigated. Time resolved plasma emission spectroscopy is coupled with a temporal shaping procedure in a closed loop. The ionic emission is enhanced relative to the neutral one via an adaptive optimization strategy. The plasma emission efficiency in case of optimized and ultrashort temporal shapes of the laser pulses are compared, evidencing an enhancement of the ionization degree of the plasma plume. Simplified temporal shapes of the femtosecond laser pulses are extracted from the optimized shape and their corresponding effect on laser induced plasma emission is discussed.

1. Introduction

Femtosecond lasers have achieved great practical interest in recent years for several applications such as precise machining, laser induced spectroscopy or biological characterization [1]. Femtosecond laser can also be used for thin film deposition of a wide variety of materials, such as diamond-like carbon [2–4], oxides [5,6], nitrides [7–9], or quasicrystals [10]. Short pulse laser irradiation has the ability to bring materials into highly non-equilibrium states and to induce rapid transformations, leading to the formation of metastable phases. Femtosecond laser ablation can drive particular thermodynamical pathways of the ablated matter close to the critical point [11,12], with influence in nanoparticles formation [13–16]. The plasma plume induced by femtosecond laser irradiation was analysed by several techniques, including time of flight analysis [2,17] or temporally resolved emission spectroscopy [18–20] and differs significantly in its kinetic properties from the plume generated by conventional ns pulses. Plasma kinetic and excitation control is a key parameter in thin films or nanoparticles synthesis, since it can improve the reactivity or the growth kinetics. A possible way to achieve higher film quality and better control of the ablation development is to regulate the interaction via spatio-temporal modification of the laser beam. Few studies were reported on the use of temporal shaped femtosecond pulses for the elaboration of thin films, mainly with double pulses for diamond-like carbon [21] or silicon carbide [22]. Further progress was made in plasma regulation for spectroscopic application [23,24] suggesting control potential by complex pulse forms. Especially for complex interaction phenomena, pulse tailoring can be connected with feedback loops. Largely applied in femtochemistry [25], adaptive optimization techniques based on real-time pulse tailoring can open large possibilities for controling the overall course of laser–matter interaction. Since prior knowledge of the interaction between laser and matter is not available, the relation between a particular temporally shaped pulse and the induced effects is not easily predictable. In that case, adaptive optimization procedures may deliver the most effective way to drive the systems in a defined state [11,26]. In the context of laser ablation, the use of adaptive pulse shaping has been recently reported for ion ejection efficiency [11,26,27] and for the generation of nanoparticles with tailored size [28]. Applications in spectroscopy and pulse characterization were suggested as well [29,30]. We report here on the possibility of tailoring the plasma plume by adaptive temporal shaping. The outcome has potential interest for thin films elaboration or nanoparticles synthesis.

We present preliminary results on the use of an adaptive feedback optimization procedure that adjust the laser temporal form for modulating plasma optical properties. This work is focused on the enhancement of the ionic emission with respect to the neutral emission lines of the plasma induced by laser irradiation of an aluminum sample.

2. Experimental setup and methods

The experimental setup is depicted in Fig. 1. A Ti:saphirre laser beam (centered at 800 nm) with 150 fs pulse duration is focused

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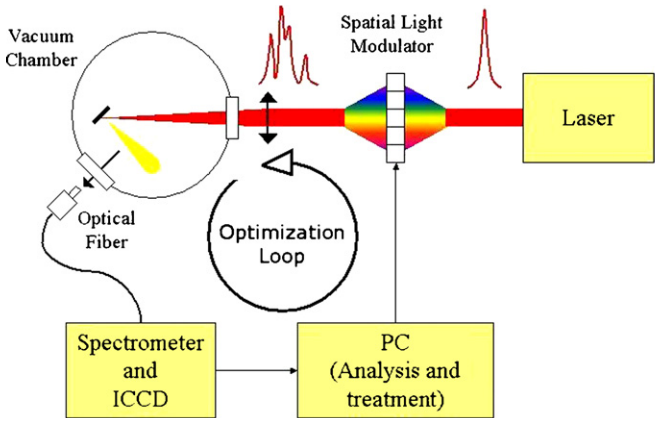

Fig. 1. Schematic description of the experimental setup.

into a vacuum chamber (~10^{-5} Pa) on pure aluminum target (99.9%), generating the ablation plasma. The fluence is controlled by precise positioning of the focusing lens and set at a value of 5.8 J/cm^2, well above the ablation threshold for Al. Prior amplification, the pulses from the femtosecond oscillator are spectrally dispersed in a zero-dispersion unit and the spatially-separated frequency components pass through a pixellated liquid crystal array acting as a Spatial Light Modulator (SLM). The device allows relative retardation of spectral components, tailoring in turn the temporal shape of the pulse [31]. The programmable nature of the SLM permits its insertion in closed loops involving rapid quantification of laser interaction, allowing thus to exercise control on the experimental results of irradiation. In this respect, plasma optical emission provides the necessary feedback by evaluating the result of the laser action.

Optical emission of the plasma plume is collected through a focusing lens with an optical fiber and injected onto the entrance slit of a spectrometer/monochromator (Chromex 500 IS/SM) equipped with two different diffraction gratings, both blazed at 400 nm (300 grooves/mm and 1200 grooves/mm). An ultrafast Intensified Charge-Coupled Device (ICCD Hamamatsu Orca 12 ER) is coupled at the output of the spectrometer, allowing a minimum temporal resolution of 3 ns. In the monochromator mode, the spectrometer is connected with a photomultiplier (Hamamatsu R-928P) and a 1 mm slit is placed before the collection lens to increase the spatial resolution. This setup permits temporal measurements of a limited spatial and spectral domain of the plasma plume. A synchronization device driven by computer is used to timely control the injection and ejection of seed pulses in and out of the amplification stages of the laser system and to trigger a delay generator (Stanford Research DG 535) which controls the camera acquisition with a temporal resolution better than 0.2 ns. All spectra reported in this article are recorded at a delay of 100 ns after the laser impact and with a time-width of 300 ns. These parameters have been chosen to maximize the acquired intensity.

This study aims to realize adaptive temporal tailoring of femtosecond pulses based on optical emission of the plasma in order to enhance the ionic population of the plasma plume with respect to the neutral one. Visible spectral lines, including the ones used to monitor ionic and neutral components of the ablated matter, are summarized in Table 1 [32,33]. We limit this study at the most intense lines. Atomic lines between 358.65 and 358.74 nm (Table 1, d) are representative of Al II population while lines at 394.40 and 396.15 nm (Table 1, e and f) give information on neutral population. The comparison of the relative intensity of these two groups of lines provides indication on the state of ionization of the ablated matter. We choose the difference between ionic and neutral lines intensity as relevant parameter (named fitness) to characterize the efficiency of a particular temporal shape to enhance the ionic population with respect to the neutral component of the plasma plume. We make the difference of ionic versus neutral lines in a given spectral range and this constitutes the quantification of the laser interaction process in a given spectral range. In order to lock up temporal shapes that maximize the fitness, we used a numerical optimization loop based on an adaptive evolutionary strategy. An initial group of arbitrary temporal shapes are chosen and tested in the experiment, while recording the irradiation results through the acquisition device. The group of corresponding spectral phase masks is ranked according to the fitness defined above. The best masks are selected, allowing to create by genetic propagators a new population that will be tested in the same way. The procedure is iteratively repeated until the convergence of the optimization loop is realized. A high number of acquisitions is needed and each acquisition is made with 15 laser shots. To ensure the stability of the experiment, the target was rotated and a new site of irradiation was sputtered before any acquisition. In all this study, temporal shapes of laser pulses are measured with a cross-correlation device. The pulse shape is determined by second order nonlinear intensity cross-

Table 1
Evolution of studied lines with femtosecond and temporally optimized pulses (the multiplication factor is given with respect to lines intensity obtained with the short pulse)

Species	Wavelength (nm)	Multiplication factor		
		Optimized	DP 10 ps	S 6 ps
Al II (a)	281.701	2.94	1.9	2.22
Al I (b)	308.215	0.54	0.44	0.56
Al I (c)	309.271 and 309.284	0.56	0.44	0.54
Al II (d)	From 358.656 to 358.745	3.24	2.79	3.17
Al I (e)	394.401	0.54	0.4	0.5
Al I (f)	396.152	0.59	0.43	0.52
Al II (g)	466.306	3.64	3.44	3.96
Al II (h)	559.330	1.99	1.76	2.07
Al II (i)	618.157 and 618.168	0.6	0.55	0.66
Al II (j)	623.334 and 623.347	2.55	2.82	2.65
Al II (k)	624.480, 624.493 and 624.510	2.93	2.97	3.67
Al II (l)	704.206	1.87	1.85	1.48
Al II (m)	705.660	2.55	1.69	1.38
All visible lines		0.65	0.51	0.61
Neutral lines		0.56	0.43	0.52
Ionic lines		2.15	1.87	2.1
Total intensity (integrated between 200 and 800 nm)		0.76	0.67	0.76