Properties of plasmas produced by short double pulse laser ablation of metals

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Abstract. We investigate the composition of plasmas produced by laser ablation of metals with two time-delayed short laser pulses using fast imaging and time- and space-resolved optical emission spectroscopy. The ablated material is deposited on mica substrates and analyzed by atomic force microscopy. The laser-produced craters are inspected by optical microscopy to evaluate the ablated material quantity. It is shown that the fraction of nanoparticles in the ablation plume is strongly altered when a second laser pulse of sufficiently large delay is applied. Comparing the results obtained for different metals, we observe a significant nanoparticle reduction for interpulse delays of the order of the characteristic time of electron-lattice thermalization. More detailed analyses show that the plume changes occur on two different characteristic times, indicating two different mechanisms at its origin. Here, we discuss the involved processes and we propose a simple and efficient technique for the measurement of electron-lattice thermalization times based on plume observations during double pulse laser ablation.

1. Introduction

Pulsed laser ablation of solid materials with intense short pulses has attracted growing interest in the last decade, since reliable pico- and femtosecond laser systems are available. With respect to nanosecond laser pulses, the interaction of short laser pulses with condensed matter is significantly different as the pulse duration is shorter than the characteristic time of energy relaxation [1, 2]. Thus, ablation occurs after the laser irradiation if short pulses are employed whereas nanosecond laser pulses interact with the plume [3-5]. The heating of the plume in the nanosecond regime leads to the generation of a strong Bremsstrahlung continuum that is undesired in analytic applications by laser-induced breakdown spectroscopy (LIBS) [6, 7]. In addition, the re-heated plume exerts a pressure on the melted layer and thus generates microscopic droplets that present the main drawback in pulsed laser deposition (PLD) of thin films using nanosecond laser sources [8]. The main advantages of material processing by short laser pulses are due to the reduction of the heat-affected zone and multiphoton absorption induced by the high laser intensity. They ensure an increased precision for micro- and nanostructuring of all materials [9, 10]. According to the minimization of thermal effects and in particular the reduction of the melting depth, short pulse laser ablation is expected to be stoechiometric and therefore promising for material analysis by LIBS [11].

Recent experimental and theoretical investigations show that small particles in the nanometer size-range present a large fraction of the ablated material [12-14]. This particularity of short pulse laser ablation was attributed to the fast heating and energy relaxation. Recent theoretical investigations [15,
propose two principal mechanisms for the nanoparticle generation during the quasi-adiabatic expansion of the plume. One consists in the transformation of matter into a liquid-gas mixture that favours the nanoparticle generation. This mechanism is so-called “phase explosion” or “thermal decomposition” [17, 18]. The “mechanical decomposition” of the metastable melt due to shock- and rarefaction waves was foreseen to present another source of efficient nanoparticle generation [19]. The particular role of pressure relaxation was stressed by Chimier and co-workers [20] and Norman et al. [21]. Nanoparticle formation by condensation is supposed to play a minor role during short pulse ablation under vacuum [22].

The precise theoretical description of short pulse laser ablation and nanoparticle generation is difficult as the knowledge of material properties is incomplete in the extreme non-equilibrium conditions that are characteristic of short pulse laser heating. Two main approaches are applied for theoretical modelling. Both are based on the two-temperature (TTM) model [23] that takes into account that electrons and lattice have different temperatures under material excitation by short laser pulses. The combination with a hydrodynamic model was employed in several numerical studies [12, 16, 24, 25] whereas the microscopic description using molecular dynamics was alternatively used in other numerical investigations [18, 19, 26, 27, 28]. The latter approach benefits from the strong enhancement of computational performance in the past years. The difficulty of the molecular dynamics calculations is due to the lack of knowledge of the interaction potential between the individual particles. In the hydrodynamic approach, the main problem comes from the incomplete knowledge of the equation of state. The improvement of both theoretical approaches requires therefore reliable experimental data characterizing the ablation of materials in the short pulse regime.

In this context, the here presented experimental investigations were mainly motivated by a deeper understanding of short pulse laser ablation and nanoparticle generation from metal targets. A particular interest was focused on the reduction of nanoparticles in the ablation plume. Nanoparticles radiate a blackbody-like continuum [22, 29, 30] and their presence is therefore not suitable for material analysis via LIBS. The elimination of nanoparticles may be also suitable for the deposition of ultra-smooth thin films via PLD [31] or for high-precision micromachining [32].

We have shown in previous papers that the fraction of nanoparticles generated during metal ablation only weakly depends on the laser fluence [14] but is strongly influenced by the strength of electron-lattice coupling [33]. This behaviour was attributed to the competition between the fast electron heat transport and the energy transfer to the lattice that governs the metals heat regime for irradiation with laser pulses shorter than the characteristic time of electron-lattice thermalization $\tau_{el}$. An increase of laser fluence favours the heat transport towards the bulk, increasing thus the ablation depth while the average thermal energy of the ablated material remains almost constant. The influence of the electron-lattice coupling was evidenced by comparing short pulse laser ablation of gold and copper. Indeed, strong coupling in the case of copper limits the heat transport and the ablation depth is accordingly small. The average energy of the ablated material is large as evidenced by the large atomization degree of the plume [33]. Contrarily, the more efficient heat transport in the weakly coupling gold leads to a larger amount of ablated material having lower average thermal energy as evidenced by the large fraction of nanoparticles within the plume.

In the present paper, we investigate the short pulse ablation of metals with two time-delayed femtosecond laser pulses.

Recently, the so-called “double pulse” technique became popular, in particular in material analysis by LIBS, where an increased analytical signal and a better signal-to-noise ratio are requested [34]. In this application, mainly performed with nanosecond laser sources, the application of the second delayed laser pulse may have different roles. In most cases, an enhanced excitation due to plasma re-heating by the second pulse is desired [35]. In some cases, the first laser pulse is applied to induce a shock wave in the surrounding air. Thus, the second laser pulse ablates the material under reduced pressure, minimizing thus the contribution of ambient air to the plasma emission [36]. Semerok and Dutouquet [37] ablated copper and aluminium with double pulses of durations ranging from 50 fs to 2 ps. They observed an increase of the plume brightness and a decrease of crater depth for delays in
the range from 1 to 10 ps. Both effects were attributed to plasma re-heating by the second laser pulse. Ablating the same metals with laser pulses of 5 ps duration, Le Harzic et al. [38] observed a reduced ablation depth and a smoother crater bottom when applying a second laser pulse with a delay of about 10\(^2\) ps. Performing plume analysis during laser ablation of titanium with double pulses of 100-fs duration, Scuderi et al. [39] observed that the amount of nanoparticles decreased with the delay between both pulses whereas the number of atoms and ions increased. They attributed the changes of plume composition to the fragmentation of nanoparticles. The double pulse ablation of silicon has been investigated by time-of-flight mass spectrometry [40] and optical emission spectroscopy [41, 42], demonstrating that the ion yield and the plume emission intensity significantly increase for delays in the range of some tens of picoseconds. In addition, the laser-produced crater becomes smoother [40]. These changes were attributed to the alteration of the optical properties that take place when the material melts. With respect to solid silicon, the melt has a much smaller optical penetration depth leading thus to a strongly increased laser energy coupling.

In metals, strong absorption occurs at any temperature due to the large number of free electrons [43]. In that case, as mentioned above, the ablation regime and thus the plume composition are determined by the interplay between electron heat diffusion and electron-lattice coupling [33]. The theoretical description requires thus the knowledge of the electron heat conductivity \(k_e\) and the electron-lattice coupling parameter \(G\) as functions of electron temperature \(T_e\) and lattice temperature \(T_l\). As rough estimations, the electron heat conductivity is given by \(k_e = k_0 T_e / T_l\), whereas \(G\) is taken constant. Here, \(k_0\) is the electron heat conductivity at ambient temperature. More precise evaluations of both parameters were recently proposed by Lin et al. [44]. In the present paper, we will limit us to qualitative interpretations of the experimental results based on the simple theoretical considerations of the electron-lattice coupling.

2. Experimental arrangement

A schematic presentation of the experimental setup is given in figure 1. The ablation experiments were carried out with a Ti:Sapphire laser (Spectraphysics, model Hurricane) delivering pulses of 100-fs duration at a repetition rate of 1 kHz. The laser was operated at 800-nm wavelength. An aperture of 2-mm diameter was employed to select the centre part of the Gaussian beam which was imaged with an achromatic lens of 50-mm focal length onto the target surface. The two delayed laser pulses were obtained by turning the beam polarization with the aid of a half-wave plate and splitting the laser beam with the aid of a polarized large band prism.
Each beam crosses a quarter-wave plate before and after reflection on a mirror at 90° incidence. After recombination by the prism, two laser pulses of equal energy and orthogonal polarization were obtained. The interpulse delay was varied from 0 to 300 ps by adjusting the length of one beam path. According to the total laser pulse energy of $2 \times 25 \, \mu \text{J}$ incident onto the samples surface and a spot diameter of 35 µm, a maximum laser fluence of $2 \times 2 \, \text{J cm}^{-2}$ was obtained. A mechanical shutter was used to control the number of applied double pulses.

The metal targets of 0.5-mm thickness, $10 \times 10$-mm$^2$ area and 99.95% purity were placed in a vacuum chamber of $10^{-4}$-Pa residual pressure. Inside the chamber, target holder and focusing lens were mounted on motorized translation axes. A glass plate was placed between target and lens to prevent the latter from deposition of ablated material. The glass plate was replaced regularly to minimize the laser beam extinction by the deposit. Fast imaging of the ablation plume was performed with the aid of a focusing objective and an intensified charge-coupled device (ICCD) (Andor, model iStar). The time delay between laser pulse and observation gate was adjusted to 400 ns with the aid of a delayed pulse generator. During the experiments, 20 laser shots were applied at maximum to each irradiation site. For measurements that require data acquisition over more than 20 ablation events, several sites were irradiated. The target was translated 50 µm perpendicularly to the laser beam to separate the irradiation sites. The laser-produced craters were inspected by optical microscopy after the ablation experiments. The ablated material was deposited on mica substrates, placed parallel to the target at a distance of 20 mm from the target surface. It was analyzed by atomic force microscopy (AFM) in the semi-contact mode. Several areas of $2 \times 2$-µm$^2$ dimension were scanned to characterize each deposit in order to count a number of particles sufficiently large for statistical analysis of the particle size distribution.

3. Results and discussion

3.1. Influence of interpulse delay on plume composition and ablation depth

In the present section, we evaluate the influence of the interpulse delay on the composition of the laser-produced plasma and on the quantity of ablated material. In addition, the plume excitation temperature is measured for copper ablation using optical emission spectroscopy.

3.1.1. Analysis of plume composition. Two plume images recorded during ablation of copper using a single pulse (a) and two delayed laser pulses (b) are shown in figure 2.

![Figure 2](image_url). Plume images recorded during ablation of copper with a single pulse of 4–J cm$^{-2}$ laser fluence (a) and with two pulses of 2 J cm$^{-2}$ delayed by 33 ps (b). The intensity profiles (c) and (d) were obtained from the images (a) and (b), respectively. The delay between observation gate and laser pulses was 400 ns.
Emission intensities of atoms (red colour) and nanoparticles (blue colour) deduced from plume images (see figure 2) for various delays between two laser pulses of 2 J cm$^{-2}$. The continuous lines present the intensity increase and decrease according to formula (1), suggesting two characteristic times for the observed plume changes. With respect to ablation of gold, the changes of emission intensity occur at shorter times for ablation of copper.

To enhance the presentation quality, the intensity matrix was smoothened by using an appropriate procedure. Both images reveal the presence of two plume components. A “slow” component of high intensity is located close to the target whereas a “fast” component of lower emission intensity is observed at larger distance. The splitting into two main plume components was observed for short pulse laser ablation of several metals [14, 22, 29, 30]. Analysis by optical emission spectroscopy revealed that the fast component mainly contains neutral atoms, whereas nanoparticles dominate the slow component. The ten-level colour palette is adjusted to the emission intensity of nanoparticles.

With respect to the single-shot experiment (a), a large increase of the emission intensity of the atomized plume component is observed when two delayed pulses were applied (b). At the same time, the nanoparticles plume intensity decreases as shown by the intensity profiles (c) and (d) that were obtained from the plume images (a) and (b), respectively, by averaging the signal over several pixel rows around the plasma symmetry axis as indicated by the dashed lines in figure 2 (a).

The emission intensities of atoms and nanoparticles deduced from the plume images are presented in figure 3 as functions of interpulse delay for copper (a) and gold (b). It is shown that strong changes of the emission intensities of atoms and nanoparticles occur for both metals with increasing interpulse delay. Indeed, the emission intensity of atoms increases by a factor of four whereas the nanoparticle intensity decreases by the same factor. With respect to ablation of gold (b), the intensity changes occur at a shorter delay for double pulse ablation of copper (a). The evolution of the emission intensity changes of both components versus interpulse duration $t$ can be approximated by the following biexponential function [45]

$$I(t) = I_0 + I_1 \left(1 - e^{-t/\tau_1}\right) + I_2 \left(1 - e^{-t/\tau_2}\right).$$

Here, $I_0$ is the intensity for an interpulse delay $t = 0$, $I_1$ and $I_2$ are the amplitudes of intensity changes that occur on different time scales, characterized by $\tau_1$ and $\tau_2$, respectively. The continuous and dashed lines present the emission intensities of atoms and the nanoparticles, respectively, calculated with formula (1) by inserting the parameters listed in table 1.
Table 1. Intensity values $I_0$, $I_1$ and $I_2$ and characteristic times $\tau_1$ and $\tau_2$ used to describe the emission intensity evolution of atoms and nanoparticles as a function of interpulse delay using formula (1).

| Metal   | Plume component | $I_0$ (a.u.) | $I_1$ (a.u.) | $I_2$ (a.u.) | $\tau_1$ (ps) | $\tau_2$ (ps) |
|---------|-----------------|-------------|-------------|-------------|---------------|---------------|
| Cu      | atoms           | 95          | 105         | 240         | 10            | 140           |
| Cu      | nanoparticles   | 117         | -78         | -12         | 13            | 200           |
| Au      | atoms           | 45          | 0           | 240         | -             | 200           |
| Au      | nanoparticles   | 1740        | -1170       | 0           | 30            | -             |

The approximation by a biexponential function suggests two characteristic times for the observed plume changes. The shorter time $\tau_1$ scales about 10 ps for copper and 30 ps for gold. These values are close to the characteristic times of electron-lattice relaxation $\tau_{el} \approx 1–10$ ps and 30–100 ps for copper and gold, respectively [46, 47]. The time $\tau_2$ is about one order of magnitude larger than $\tau_1$. It characterizes a slower process that strongly influences the atomic emission intensity increase as shown by the relative large $I_2$-value. Contrarily, we have $I_2 / I_1 << 1$ for the nanoparticle emission intensity evolution, indicating thus that the slow process has no significant influence on this component of the plume.

The reduction of the amount of nanoparticles with increasing interpulse delay was confirmed by atomic force microscopic analyses of the ablated material deposited on mica substrates. To illustrate the strong decrease of the number of deposited nanoparticles with $t$, we present in figure 4 AFM images of mica substrates after deposition with 500 laser double pulses and three different interpulse delays. With respect to ablation with $t = 0$ ps (a), the number of nanoparticles is strongly reduced when increasing the interpulse delay to 6 ps (b). A further decrease occurs for long $t$-values, as illustrated in figure 4(c), where only a few nanoparticles are observed. The number of nanoparticles deduced from the AFM analysis is presented in figure 5 as a function of interpulse delay and compared to the biexponential function [equation (1)] that was used to characterize the nanoparticle emission intensity during copper ablation (see figure 3).

3.1.2. Ablation depth measurements. The ablation depth was measured in the following way: for each interpulse delay, a series of 10 craters was drilled with an increasing number of double pulses as shown in figure 5. The depth of each crater was measured using optical microscopy by focussing on the samples surface and the crater bottom [48]. According to the linear increase of crater depth $z$ with the laser pulse number $n_{laser}$, the ablation depth was deduced from the slope $\Delta z / \Delta n_{laser}$.
The ablation depth as function of interpulse delay is presented in figure 6 for ablation of copper and gold. For an interpulse delay \( t = 0 \), the ablation depth equals the value obtained with a single pulse of \( 4 \text{ J cm}^{-2} \) fluence, whereas the depth measured for large delays is slightly smaller than the value corresponding to one pulse of \( 2 \text{ J cm}^{-2} \) fluence. This behaviour is in agreement with the ablation depth measurements performed by Semerok and Dutouquet [36] after laser ablation of copper with short laser pulses of durations ranging from 50 fs to 2 ps. Similar to the characterization of the plume emission intensities, we approximate the ablation depth by a biexponential function by replacing the intensity values \( I \) in equation (1) by the appropriate depths \( z \) (continuous and dashed lines for copper and gold, respectively). It is noted, that the times \( \tau_1 \) and \( \tau_2 \) equal the characteristic times of the plume intensity evolution. The main contribution of the observed ablation depth decrease is attributed to the change of the samples heat regime as we have \( z_1 >> z_2 \), similar to the evolution of nanoparticle emission intensity. This indicates that nanoparticles present the major part of the ablated mass.
Excitation temperature measurements. The excitation temperature of atoms was determined for double pulse laser ablation of copper from the intensity ratio of the Cu I 510.55 and 515.32 nm transitions having significantly different upper level energies (see table 2). Both lines are observed in the spectra presented in figure 7 for different interpulse delays. It is shown that the Cu I 515.32 nm-intensity significantly increases with $t$, whereas the Cu I 510.55 nm-intensity is almost constant.

Table 2. Radiative decay $A$, upper level energy $E$ and statistical weight $g$ of the spectral lines used for temperature measurements.

| Transition      | $A$ (s$^{-1}$) | $E$ (cm$^{-1}$) | $g$ |
|-----------------|----------------|----------------|-----|
| Cu I 510.55 nm  | $2.0 \times 10^6$ | 30784          | 4   |
| Cu I 515.32 nm  | $6.0 \times 10^7$ | 49935          | 4   |

Assuming a Boltzmann distribution of the excited level population densities, the temperature can be deduced from the intensity ratio of spectral lines emitted from the same species using

$$T = \frac{E_2 - E_1}{k_B} \left[ \log \left( \frac{I_1 \lambda_1 A_1 g_1}{I_2 \lambda_2 A_2 g_2} \right) \right]^{-1}. \quad (2)$$

Here, $k_B$ is the Boltzmann constant, $\lambda$ and $I$ are the resonance wavelength and the measured intensity of the transitions, respectively. The indexes 1 and 2 stand for the transition of larger and smaller upper level energy (see table 2). The derived excitation temperature is presented in figure 8 versus interpulse delay. It is shown that the temperature is almost constant up to an interpulse delay of about 20 ps and a significant increase is only observed for larger $t$-values. The late temperature increase is in agreement with the observed intensity rise of the atomized plume component (section 3.1.1) that was described by the larger characteristic time $\tau_2$ in the biexponential function [equation (1)]. The delay of the increase supports the assumption that plasma heating due to the interaction of the second laser pulse with the vaporized matter is an additional mechanism responsible of the plume alteration, different from processes that cause the nanoparticle reduction on the timescale of electron lattice relaxation.
3.2. Probing electron-lattice coupling via plume observations

In the previous section, we demonstrated that the plume changes may be characterized by two different times. The shorter time equals the characteristic time of electron-lattice thermalization and describes the most significant change of plume atomization. In the present section, we take advantage of the dependence of the plume changes versus interpulse delay in order to deduce the characteristic time of electron-lattice thermalization of transition metals. Thus, we focus our attention on the plume changes that occur on the shorter timescale characterized by $\tau_1$. At this timescale, the emission intensity variation versus interpulse delay is characterized by a monoexponential function $I(t) = I_0 + I_1(1 - e^{-t/\tau_1})$. Here, $I_0$ is the intensity at $t = 0$, $I_0 + I_1$ the intensity at $t = \infty$, and $\tau_1$ the characteristic time of plume intensity change that equals the electron lattice thermalization time. After the evaluation of $I_0$ and $I_1$ and assuming $\tau_{e-l} = \tau_1$, the characteristic time of electron lattice thermalization can be derived from the linearized function [49]

$$\log[1 - (I - I_0)/I_1] = t/\tau_{e-l}.$$  \hfill (3)

**Table 3.** Atomic mass $M$, characteristic time electron-lattice relaxation $\tau_{e-l}$ deduced from the atomic emission intensity changes versus interpulse delay according to equation (3) for double pulses with two different laser fluences.

| Metal | $M$ (amu) | $\tau_{e-l}$ (ps) $2 \times 0.5$ J cm$^{-1}$ | $\tau_{e-l}$ (ps) $2 \times 2$ J cm$^{-1}$ |
|-------|-----------|---------------------------------------------|---------------------------------------------|
| Ti    | 47.9      | 50                                          | 60                                          |
| Zr    | 91.2      | 90                                          | 110                                         |
| Hf    | 178.5     | 200                                         | 240                                         |

Applying equation (3) to the atomic emission intensities measured for Ti, Zr and Hf, a linear evolution is observed for the three metals with different slopes as shown in figure 9. It is noted that the inferred $\tau_{e-l}$-values follow the atomic mass ratio 1:2:4 of the three metals (see table 3). The linear increase of $\tau_{e-l}$ with the atomic mass $M$ is expected. At large electron temperature, the metal is expected to behave like a hot plasma and the electron-lattice relaxation scales as $\tau_{e-l} \propto M T_e^{3/2}$ [5].

![Figure 9. Analysis of the atomic emission intensity dependence versus interpulse according to equation (3) for ablation of Ti, Zr and Hf with double pulses of $2 \times 0.5$ J cm$^{-2}$.](image)
An almost linear dependence of $\tau_{e-l}$ on $M$ was also predicted by the calculations of Eidmann et al. for Cu, Ag, and Au [24].

Beside the $\tau_{e-l} \propto M$ dependence, we observe a slight influence of the laser fluence on the electron-lattice thermalization. The electron-lattice thermalization times listed in table 3 show that an increase of laser fluence by a factor of 4 (from $2 \times 0.5$ to $2 \times 2$ J cm$^{-2}$) leads to a $\tau_{e-l}$-augmentation of about 20% only. The formula presented above predicts a strong increase of the relaxation time in $T_{e}^{\text{rel}}$ that is a consequence of the lowering of the energy exchange rate with increasing electron velocity. We deduce that $T_{e}$ only weakly increases with laser fluence, and that most additional laser energy leads to an enhanced energy transport into the bulk [14, 33].

A broad spread in the values of electron lattice relaxation time and coupling parameter can be observed in literature. In case of nickel, for instance, $G$-values ranging from $3 \times 10^{17}$ to $10^{17}$ W m$^{-3}$K$^{-1}$ have been published [50, 51]. The data were obtained using different values of laser fluence. Most $\tau_{e-l}$ and $G$ data were obtained in pump-probe experiments by measuring changes in surface reflectivity [52, 53]. These experiments are limited to relatively weak laser excitation to avoid nonlinear saturation processes occurring in very thin metal films ($\approx 200$ nm) used as targets. Relaxation times ranging from 1 to 4 ps and larger coupling parameters of about $10^{17}$ W m$^{-3}$K$^{-1}$ were reported for copper [52]. Corkum et al. [54], however, studied multipulse laser-induced damage to deduce the surface temperature of bulk metal targets. These authors obtained $G$-values one order of magnitude smaller than the values reported in references [52] and [55] for the same material. The $\tau_{e-l}$ values we present in table 3 are therefore consistent with the values reported in literature.

4. Summary and conclusion
The present experimental investigations of metal ablation with two time-delayed short laser pulses demonstrate that the plume composition is strongly altered if the interpulse delay is of the order of the electron-lattice thermalization time. The changes of the emission intensities of atoms and nanoparticles with the interpulse delay reveal two characteristic times, suggesting different mechanisms to be responsible for the plume alteration. The samples heating regime is expected to be responsible for changes on the timescale of electron-lattice thermalization. With increasing lattice temperature, the electron heat conductivity decreases and the laser energy deposited by the second laser pulse is confined in a smaller volume. According to the decrease of the heated depth and the rise of the average temperature of the ablated matter, the atomization degree of the plume increases. Effects of pressure relaxation are also expected to cause plume changes on the timescale of electron-lattice thermalization [20, 21]. For times about one order of magnitude larger, plasma re-heating due to interaction of the second delayed laser pulse with the vaporized material is expected. The observations are supported by measurements of the ablation crater depth as well as AFM analysis of the nanoparticles deposited on mica substrates. The present experiments indicate that the change of the samples heat regime is the dominant mechanisms for the reduction of nanoparticles within the plume, whereas the increase of the atom emission intensity is mainly due to plasma re-heating by the second delayed laser pulse.

In addition, we found for Ti, Zr and Hf that the electron-lattice thermalization time deduced from the plume changes increases linearly with the atomic mass as expected for the electron-ion thermalization in a hot plasma. Furthermore, $\tau_{e-l}$ is shown to increase slightly with laser fluence as a result of the change of the heat regime. The present investigations demonstrate the possibility of characterizing the electron-lattice coupling of metals by simply observing the ablation plumes produced laser ablation with two delayed short laser pulses. With respect to conventional pump-probe reflectivity measurements, the presented results demonstrate that multipulse laser irradiation can be applied for measurements of the electron-phonon coupling.

Finally, the efficient reduction of the amount of nanoparticles within the plume on one hand, and the increase of the atomic emission intensity on the other hand, improve the performances of material analysis via LIBS. The double-pulse technique may be also promising for other applications such as micromachining or pulsed laser deposition.
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