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Thomson scattering from laser induced plasma in air

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Abstract. The laser induced plasma in air produced by 6 ns, 532 nm Nd:YAG pulses with 25 mJ energy was studied using the Thomson scattering method and plasma imaging techniques. Plasma images and Thomson scattered spectra were registered at delay times ranging from 150 ns to 1 \( \mu \) s after the breakdown pulses. The electron density and temperature, as determined in the core of the plasma plume, were found to decrease from \( 7.4 \times 10^{17} \) cm\(^{-3}\) to about \( 1.03 \times 10^{17} \) cm\(^{-3}\) and from 100 900 K to 22 700 K. The highly elevated electron temperatures are the result of plasma heating by the second, probe pulse in the Thomson scattering experiments.

1. Introduction

Laser induced breakdown spectroscopy (LIBS) is a well known and commonly used technique for analysis of solid, liquid and gaseous samples [1, 2]. However, it suffers from low sensitivity and poor detection limits when compared to other spectroscopic methods [3]. Increasing the sensitivity of LIBS would also allow one to decrease the pulse energies and minimize destructive effects on the sample. It was recently demonstrated that double pulse (DP) LIBS is an effective way to improve the analytical capabilities of the LIBS method [4]. Higher sensitivity is achieved by enlarged plasma volume, more efficient production of atoms in excited states and longer sustained emission. Although the mechanism resulting in signal enhancement in DP LIBS is not fully recognized, lowering of the laser shielding effect, heating of the target surface and reheating of the formed plasma plume have been invoked as possible explanations of the experimental observations. DP LIBS has been realized in orthogonal and parallel configurations of the laser beams using either the pre-ablation or the re-heating mode [4]. In the orthogonal re-heating configuration, the first pulse irradiates the sample and generates the plasma plume while the second pulse, propagating orthogonally to the sample and to the first laser beam, re-heats the ablated elements resulting in higher plasma emissivity.

In this configuration, the impact of the second laser pulse on the plasma plume can be studied using standard emission spectroscopy, but also by analyzing its scattering by plasma electrons in the Thomson scattering (TS) experiment. Unlike emission spectroscopy, the principal advantages of TS are high spatial resolution and ease with which the measured data can be interpreted. The standard plasma parameters, the electron temperature and density, can be
directly derived from the electron feature of the TS spectrum without any assumptions about the plasma symmetry, equilibrium state or its chemical composition, whereas the composition itself can be deduced from the ion feature. Detailed theory and applications of TS can be found in several monographs [5, 6].

In this work we used TS to investigate double-pulse laser induced plasma in air, since most of LIBS experiments with solid samples are performed in such an environment.

2. Experimental arrangement
The experiments were performed in atmospheric pressure air with two second-harmonic (532nm) Nd:YAG laser beams arranged in orthogonal geometry. The first pulse induced the plasma plume while the second, with much lower fluence and below the breakdown threshold for air, was used simultaneously to reheat the plasma and for observation of Thomson scattering. These beams were delivered to the interaction region and focused to the spots of 30 and 150 μm diameters using lenses of 10 cm and 50 cm focal length, respectively. The energy of the laser pulses was 25 mJ and 50 mJ, respectively and their duration was about 6 ns. This way, the fluences of the laser pulses in the interaction region differed by more than one order of magnitude. The delay between laser pulses was controlled by a delay generator and monitored using a fast photodiode and an oscilloscope.

The image of the induced plasma and the Thomson scattered light were observed in the perpendicular direction to the laser beams (see insert in Fig. 2), by imaging the investigated plasma volume onto the entrance slit of a spectrograph with 1.6 nm/mm reciprocal dispersion. The TS spectra were collected over a wavelength range of about 20 nm, with the slit width set to 50 μm. Then the images of the plasma were recorded at zero order of the spectrograph with the slit fully opened to 3.8 mm. The spectral and spatial resolution were found to be 0.032 nm and 31 μm, respectively. Single-shot TS spectra and plasma images were recorded using a gated two-dimensional intensified charge-coupled device (ICCD) camera and averaged over 1000 shots. The ICCD was synchronized either to the first (plasma imaging) or to the second (TS measurements) laser pulse and its integration time was set to 8 ns and 15 ns, respectively.

3. Results and discussion
Series of images of the plasma induced by the first laser pulse were recorded from 10 ns to 1000 ns after the pulse. Since measured intensities are integrated along the line of sight, the spatial distribution of the total emission coefficient was retrieved by applying the Abel transformation. Typical images of the plasma plume after Abel transformation are presented in Fig. 1a, where the intensity in each individual picture is normalized to the maximum value. The evolution of

![Figure 1](image)

Figure 1. (a) Temporal evolution of Abel inverted images of the plasma plume (an air spark). The timings in the images represent the time after the breakdown pulse. The exposure time was 8 ns. (b) Temporal evolution of plasma emission coefficient taken from the images at the central part (z = 0) of the image and at the y position marked by the line is shown in Fig. 1b. Rapid radial and axial expansion of the plasma
plume occurs during the first 200 ns, while the axial expansion is predominantly in the direction of the propagating laser beam. It can be seen here that the plasma expansion can be related to fast decay of plasma emissivity (mainly due to the bremsstrahlung process) on the axis due to the decrease of the electron density caused in turn by recombination processes. For longer times, spatial evolution becomes more complex and the plasma size remains nearly constant, which corresponds to a smaller decay rate of its emissivity.

Images of the TS spectra as registered by the ICCD camera are shown in Fig. 2. They were obtained by illuminating the central part of plasma plume with the second laser beam and along the line marked in Fig. 1: the $z$ axis at $y = 0$. The consecutive images refer to different time delays between laser pulses. The saturated part of the spectra near 532 nm, apart from the TS (ion feature) and plasma light, contains also the Rayleigh scattered light as well as stray light and was discarded from further analysis. On the other hand, the electron feature appears as two peaks symmetrically situated with respect to the laser wavelength indicating the partially collective character of TS. This part of the TS spectrum was observed, with reasonable signal-to-noise ratio, for time delays longer than 150 ns. For shorter delays, the electron feature gets very broad and disappears in the midst of very strong continuous emission of plasma. For time delays shorter than 600 ns, due to large separation between peaks, only one side of the TS spectra could be recorded at the same time. The collected TS spectra extend spatially over a distance which corresponds to plasma diameter. The spectra in Fig. 2 clearly reveal that the

![Figure 2](image)

**Figure 2.** Thomson scattered spectra by laser induced plasma taken for different delays between laser pulses. The ICCD exposure time was 15 ns. The plasma generating beam is applied along the $y$-axis, the probe beam along $z$-axis while scattered light is observed in the $x$ direction. Spectra of scattered light were taken from regions around the core of the plasma ($x = y = 0$).

![Figure 3](image)

**Figure 3.** Example of the measured and fitted Thomson scattered spectra. The consecutive figures correspond to 150 (a), 400 (b) and 1000 ns (c) delay time between the breakdown and TS probe laser pulses. The fitted $n_e$ and $T_e$ are $7.44 \times 10^{17}$ cm$^{-3}$ and 100 900 K (a), $3.35 \times 10^{17}$ cm$^{-3}$ and 45 900 K (b) and $1.03 \times 10^{17}$ cm$^{-3}$ and 22 700 K (c).

peaks diverge and become broader approaching the origin of the plasma, meaning that both $n_e$ and $T_e$ are increasing. For more quantitative analysis theoretical TS spectra were fitted to the experimental ones as described in [8]. Such theoretical and experimental spectra are shown in
Figure 3 for time delays of 150, 400 and 1000 ns. The fitted $n_e$ and $T_e$ depending on time delay between laser pulses are depicted in Fig. 4. The electron density decreases with delay time from about $7.4 \times 10^{17}$ cm$^{-3}$ to about $1.03 \times 10^{17}$ cm$^{-3}$. At the same time the electron temperature drops from 100 900 K to about 22 700 K. It should be emphasized that the TS spectra are integrated over the duration of the second (probe) pulse, which heats the plasma in the inverse bremsstrahlung process. The problem of thermal plasma heating by the laser pulse and its influence on the TS results was recently studied theoretically by Murphy [7] and experimentally by Dzierżęga et al [8]. It was shown that application of the laser pulse results (under conditions of typical TS experiments) in the increase of $T_e$. On the opposite, the change of $n_e$ is insignificant due to the long ionization time scale with respect to the duration of applied laser pulses. Thus, the electron temperature derived from TS spectra can significantly overestimate that just before the probe pulse. On the other hand, the electron density, at the first approximation, can be assumed to be unaffected. This means that derived values of $T_e$ and $n_e$ are close to those at the end of the second pulse, not to the unperturbed ones. Moreover, since the process of inverse bremsstrahlung is proportional to $n_e^2/\sqrt{T_e}$, the increase of $T_e$ is expected to be higher when the plasma is probed by the TS pulse at the early stages of its evolution.

In a future experiment, plasma heating during the probe TS pulse will be studied in order to determine the initial "undisturbed" temperature of the plasma generated by the first pulse.

Acknowledgments
We wish to acknowledge the support of this work by project d’Action Intégrées – Polonium 7836/R09/R10,7835/R09/R10 and partially by the Polish Ministry of Science and Higher Education grant NN202 031136.

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