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Simultaneous measurement of electron and heavy particle temperatures in He laser-induced plasma by Thomson and Rayleigh scattering

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Thomson and Rayleigh scattering methods were applied to quantify the electron and heavy particle temperatures, as well as electron number density, in a laser spark in helium at atmospheric pressure. Plasma was created using 4.5 ns, 25 mJ pulses from Nd:YAG laser at 532 nm. Measurements, performed for the time interval between 20 ns and 800 ns after breakdown, show electron density and temperature to decrease from $7.8 \times 10^{23}$ m$^{-3}$ to $2.6 \times 10^{22}$ m$^{-3}$ and from 95 900 K to 10 350 K, respectively. At the same time, the heavy particle temperature drops from only 47 000 K down to 4100 K which indicates a two temperature plasma out of local isothermal equilibrium.

Laser-induced plasmas (LIP) have found numerous applications, including laser ablation, short wavelength sources for lithography, micromachining as well as the qualitative and quantitative elemental analysis of different kinds of samples. A detailed description of plasma, important for modeling and analytical purposes, requires a thorough knowledge of atom, ion, and free electron number densities and their temperatures. These parameters are commonly deduced in an indirect way from optical emission measurements, assuming plasma in local thermodynamic equilibrium (LTE) which is often very questionable. The concept of LTE has various aspects which are discussed in detail by van der Mullen and recalled by Cristoforetti et al. for the case of LIP. For example, two balances—isothersmal and Saha-Boltzmann—should be locally in equilibrium, which is difficult to verify using such a simple criterion as the McWhirter one.

The purpose of the current work is to expand on our recent experiments in LIP and to demonstrate how in such plasma, electron and heavy particle temperatures and the electron number density can be simultaneously determined from laser scattering spectra. In this way, the local isothermal equilibrium can be validated.

Our experimental setup is depicted in Fig. 1. A vacuum chamber was evacuated below 0.1 mbar and then purged with helium at atmospheric pressure at a flow rate of 70 l/h. Plasma was created in the center of the chamber by focusing Nd:YAG laser pulses (4.5 ns, 25 mJ at 532 nm and $\Delta \lambda \approx 28$ pm) with an 80 mm focal length lens. The laser spots of 44 $\mu$m full width at half maximum (FWHM) resulted in a laser fluence of about 1.1 kJ/cm$^2$, and shot to shot highly reproducible plasma plumes. For laser scattering, a separate, single-mode ($\Delta \lambda < 0.28$ pm), Nd:YAG laser with pulse duration of about 6.0 ns and energy of 20 mJ at 532 nm was used. The beam was directed perpendicularly to the breakdown pulse and loosely focused to the spot of 440 $\mu$m FWHM by means of 500 $\mu$m focal length lens resulting in energy fluence of 9.1 J/cm$^2$. The emission from LIP and the scattered light were observed at 90° with respect to the directions of the laser beams by imaging the detected light onto the entrance slit of a spectrograph (Acton SP-2750) with 750 mm focal length. The spectrograph was equipped with 2 gratings of 1200 gr/mm and 2400 gr/mm and of 1.005 nm/mm and 0.502 nm/mm reciprocal dispersion, respectively. The images were magnified by a factor of 1.2 by the collecting optics. In order to minimize the back scattered laser light, the viewports were mounted at the Brewster angle at the end of the chamber tubes, at a long distance from the plasma.

Plume imaging was accomplished at the zeroth order of the spectrograph with the slit fully opened, i.e., up to 3.0 mm. The plume images and the laser scattered spectra were recorded using a gated intensified CCD (ICCD) camera and were averaged over 100 (plume image) or 5000 (laser scattering) laser pulses with the ICCD gate width set to 8 ns. The two lasers and the ICCD were synchronized using a delay generator.

The total power scattered by our plasma per unit frequency $d\omega$ and within a solid angle $d\Omega$ can be given as

$$d\omega = P_L L \Delta \Omega \left( S_T(k, \omega) n_x \frac{d\sigma_T}{d\Omega} + S_R(k, \omega) \sum_j n_j \frac{d\sigma_R}{d\Omega} \right) d\omega,$$

where $P_L$ is the incident laser power and $L$ is the length of the detection volume along the incident laser beam. The two terms in the brackets represent contributions from scattering

FIG. 1. Scheme of the experimental setup.
The shape of the scattering spectra is described by the spectral density functions $S_T(k, \omega)$ and $S_R(k, \omega)$, where $k$ is the scattering vector and $\omega$ is measured with respect to the laser frequency $\omega_L$. For Thomson scattering, $S_T$ depends not only on electron density and temperature but also on ion temperature and the ion charge number $Z$. This spectrum can be calculated according to Evans and Katzenstein\cite{Katzenstein} or Froula et al.\cite{Froula}. In the case of a single element plasma and assuming a two-temperature plasma model, the density function for Rayleigh scattering is expressed as

$$S_R(k, \omega) = \frac{4\pi}{\Delta k_R} \frac{\ln^2}{\pi} \exp\left(-4 \ln 2 \frac{(\omega - \omega_R)^2}{\Delta \omega_R^2}\right).$$

$$\Delta \omega_R = 2\omega_L \sin(\theta/2) \sqrt{\frac{8\mu kT_e}{m_e}}.$$

(2)

where $T_e$ and $m_e$ are the atom/ion temperature and the mass of He atom, respectively. In general, the full spectral range of TS is two orders of magnitude larger than RS. Although the central ionic feature of the TS spectrum is of similar width to the RS one, it takes a different shape, strongly dependent on the ratio of $T_e$ to $T_h$, enabling decomposition of these spectra.

In Figs. 2(a)–2(c), we present LIP images recorded at different delays after the breakdown pulse. Images of the corresponding scattering spectra are shown in Figs. 2(d)–2(i). These spectra were collected while illuminating the center of the plasma plume with the probe laser, as marked in Figs. 2(a)–2(c). The TS spectrum, with distinct electron features, reveals the collective character and fitting $S_T$ yields both electron density and electron temperature. The central part of the spectrum includes the ionic feature of TS superimposed on the RS on plasma heavy particles—atoms and ions in their ground and excited states. This central part of the scattering spectrum was separately measured with maximal spectral resolution ($\Delta \lambda = 0.027$ nm with 2400 gr/mm grating) and its images are shown in Figs. 2(g)–2(i). At delays shorter than about 500 ns, this signal is much more intense than the scattering on the cold ambient He gas, outside the plasma plume. After a few hundreds of nanoseconds, these two signals become comparable and are of similar spectral width dominated by the apparatus profile.

Figure 3 shows experimental scattering spectra obtained on the axis of the plasma plume and the fitted spectral density function $S_T$ yielding the electron density and temperature. The central part of the scattering spectrum was then fitted according to Eq. (1) at given $n_e$ and $T_e$ with $T_h$ and the RS amplitude as the fitted parameters. The fitting was performed including the apparatus profile, determined from the RS spectral profile in cold He. This profile was approximated by the pseudo Voigt function with FWHM of 0.028 nm. The whole procedure of plasma diagnostics is applicable till the width of the scattering spectrum exceeds the apparatus profile which was valid for delays shorter than 1 µs.

The temporally resolved $n_e$, $T_e$, and $T_h$ are presented in Fig. 4. The electron density decreases from $7.8 \times 10^{21}$ m$^{-3}$ at 20 ns to $2.6 \times 10^{20}$ m$^{-3}$ at 800 ns after the breakdown pulse. In the same time, the measured electron temperature drops from about 95,900 K to 10,350 K and largely exceeds the heavy particle temperature which decreases from 47,000 K to 4100 K. The statistical uncertainties of $n_e$, $T_e$, and $T_h$ do not exceed 1.2%, 2.8%, and 11%, respectively. The uncertainty of the electron temperature can be additionally enlarged due to possible electron heating by the probe pulse. However, this effect was verified to be negligible and under our experimental conditions, calculations (see Mendys et al.)\cite{Mendys} indicate at most 2% increase of $T_e$. The accuracy of the presented results can be further reduced for short delays due to inevitable time and space integration of the scattered data over a quickly decaying plasma with radial dimensions comparable with the width of the probe beam.

In the studied temporal range of plasma evolution, there exists large discrepancy between the electron and heavy particle temperatures, which indicates a two-temperature plasma.
out of local isothermal equilibrium. Moreover, the measured electron densities are too low to satisfy the McWhirter criterion\textsuperscript{2,3} which needs $n_e > 10^{24}$ m\textsuperscript{-3} for our He LIP to be in the LTE. This criterion, commonly used in the literature, estimates the minimal electron density required for collisional processes to significantly dominate over radiative ones and ensure negligible deviations from the LTE. It also defines the necessary condition for establishing local Saha-Boltzmann equilibrium in homogenous and stationary plasmas.\textsuperscript{2} Therefore, we can state that our plasma is out of the LTE since neither isothermal nor Saha-Boltzmann balances are equilibrated.

Similar values of $n_e$ and $T_e$ have been recently obtained by Nedanovska et al.\textsuperscript{9} They also used Thomson scattering method for helium atmospheric pressure LIP but their results concern longer delay times, i.e., from 400 ns up to 17.5 $\mu$s.

The decays of measured plasma parameters with time were then fitted with power laws. We found $n_e$ to fall off as $t^{-0.9}$ whereas temperatures drop at very similar rates proportionally to $t^{-0.6}$. The decay rate of $n_e$ is directly related to the rate of the plume expansion which significantly affects particles’ densities and to the electronic recombination rates. On the other hand, the temperature decay is predominantly governed by the plume expansion and radiative losses due to continuum emission. However, this rate is considerably decelerated by plasma heating in three body recombination processes, when on average 24.5 eV is released by He ions.

In summary, we have used optical Thomson and Rayleigh scattering to quantify the electron and heavy particle temperatures and electron density in laser-induced plasma in helium at 1 atm. We found a large discrepancy between $T_e$ and $T_h$, indicating considerable deviations from the isothermal equilibrium in this plasma for the studied time intervals. This finding is consistent with the measured electron densities, which appear too low even to satisfy the McWhirter criterion.

We consider the presented laser scattering method very useful in LIP studies, because it is of high temporal and spatial resolution, but also since all of its results do not rely on assumptions about the equilibrium state of the plasma. Nevertheless, its wider application to different LIP—containing elements heavier than He—calls for optical detection systems of much higher spectral resolution.

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