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Absorption tomography of laser induced plasmas

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ABSTRACT

An emission tomography of laser-induced plasmas employed in the laser induced breakdown spectroscopy (LIBS) requires signal integration times in a microsecond range during which the LIBS plasma cannot be considered stationary. Consequently, the use of the data for reconstructing the plasma properties under the assumption that the latter does not change significantly during the integration time leads to inaccurate results. To reduce the integration time, it is proposed to measure a plasma absorption in parallel rays using a scanning rectangular aperture whose dimension Δ along the scanning direction is about a characteristic size of plasma plumes ($\Delta \sim 1$ cm) and the other dimension Δ_p is of the order of a uniformity length of plasma parameters ($\Delta_p \sim 10\ \mu$m). The aperture is moved step by step along the scanning direction and the total energy of photons coming through the aperture is measured during time T at each position of the aperture. Owing to the large size of the aperture, the integration time T is reduced by a factor $\sim \Delta_p/\Delta$. A numerical data processing is proposed to restore the spatial resolution of the plasma absorption along the scanning direction. It is determined by the scanning step $\Delta_s \le \Delta_p$. Another advantage of the proposed procedure is that inexpensive linear CCD or non-discrete (PMT, photodiode) detectors can be used instead of costly 2-dimensional detectors.

1. Large apertures and the Abel inversion

Let a laser pulse impinging a target create a plasma plume. Suppose that the laser ablation is such that the plasma plume is axially symmetric. In this case, the symmetry axis coincides with the laser ray. Let the coordinate system be set so that the symmetry axis is the z-axis. Then the emissivity is a function $\varepsilon = \varepsilon(r,z,t,\nu)$ where $r = (y^2+x^2)^{1/2}$, t is the time, and ν is the frequency. A measured quantity is the intensity $I(y,z,t,\nu)$ of light (or the energy flux per unit time) per unit frequency observed along the rays through an infinitesimal area element $\Delta A = \Delta y \Delta z$ centered at the point $(0,y,z)$ that propagate in a narrow solid angle $\Delta\Omega$ about the line parallel to the x-axis [1]

$$I(y,z,t,\nu) = \Delta A \Delta\Omega \int_{-\infty}^{\infty} dx \varepsilon(\sqrt{y^2+x^2},z,t,\nu)$$
$$= 2\Delta A \Delta\Omega \int_{y}^{\infty} \frac{dr r\varepsilon(r,z,t,\nu)}{\sqrt{r^2-y^2}}. \quad (1)$$

If the cross section of a plasma plume by the plane parallel xy plane (i.e. at a fixed z) is a disk of radius R, then the limits of integration in the first integral of Eq. (1) are reduced to $-R \le x \le R$ and to $y \le r \le R$ in the second integral, while y ranges over the interval $0 \le y \le R$ owing to the symmetry of the intensity relative to the reflection $y \to -y$. It is well-known that Eq. (1) can be solved for $\varepsilon(r,z,t)$ by the Abel inversion [2,3]. The equation applies to the measured intensity data only if ΔA and $\Delta\Omega$ are infinitesimal and the plasma has the axial symmetry. The smallness of ΔA and $\Delta\Omega$ guarantees that the intensity

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data are obtained only for parallel rays (contributions of angled rays are suppressed by higher powers of $\Delta A \Delta \Omega$). The axial symmetry implies that the emissivity depends only on the combination $r = \sqrt{x^2+y^2}$. If the plasma is not axially symmetric, Eq. (1) is not valid and the Abel inversion is not applicable. In this case, a reconstruction of the emissivity can be carried out by the Radon transform method [4].

In LIBS emission experiments, the smallness of ΔA is provided by a narrow spectrometer slit and small pixel size of a detector, both in the order of 10 μm, while the solid angle $\Delta\Omega$ is restricted by a low acceptance angle of a spectrometer with a high *f*-number. For a spectrometer with $f \geq 6$, $\Delta\Omega \leq 0.02$ sr. The smaller the factor $\Delta A \Delta \Omega$, the higher spatial resolution can be achieved, and the more accurate reconstruction of the emissivity can be obtained by the Abel inversion. The energy of photons collected by the detector along a given line of sight is $E = \int_0^T dtI(y,z,t,v)$. Therefore an increase of the spatial resolution as well as the accuracy of the Abel inversion necessarily leads to increasing the integration time T in order to keep the same signal-to-noise ratio in the data, when measuring E. A typical integration time in LIBS plasma experiments required for accurate plasma diagnostics that are based on the Abel inversion is $T \sim 1$ μs or even higher. If the plasma is not stationary during the time T, the collected data correspond to a time-averaged intensity and the reconstructed emissivity may not be accurate. This is especially relevant for studying elemental contents of plasmas expanding into an ambient gas because of percolation processes at a rapidly moving plasma–gas interface. Numerical simulations of LIBS plasma dynamics show that this is often the case, especially for earlier stages of the plasma evolution [5–7].

Thus, the integration time can only be reduced by increasing the geometrical factor $\Delta A \Delta \Omega$. If $\Delta\Omega$ becomes large, then photons propagating along angled rays will be collected and, hence, the approximation of parallel rays becomes invalid. This is indeed true if the plasma *emission* is collected by the slit. The problem can be avoided if the collimated light *absorbed* by the plasma is measured. Assuming that the plasma is in a local thermodynamic equilibrium, the emissivity and absorptivity are equal, which is true a few hundreds of nanoseconds after the initiation of the plasma [8]. It is therefore proposed here to measure the absorption of a collimated (or laser) light passing through the plasma plume. Owing to the assumption of the local thermodynamic equilibrium, no distinction between the terms "emissivity" and "absorptivity" is made in what follows. In particular, the geometrical factor in (1) equals the area of the aperture, $\Delta A \Delta \Omega \rightarrow \Delta A$, if the absorption is measured because angled rays are simply absent (or their contribution can be neglected as the rays coming from a collimated source or laser are (nearly) parallel). An experimental setup to measure the plasma absorption is presented in Section 2 (see Fig. 1). It is argued there in that the absorption in parallel rays can be accurately measured, provided the collecting aperture is positioned at a sufficiently large distance ρ from the plasma plume. The smaller the factor $\Delta A/(4\pi\rho^2)$, the higher is the accuracy of absorption measurements.

In the absorption experiment, the integration time can therefore be reduced by increasing the aperture area ΔA. Consider a rectangular aperture that has an infinitesimal height Δz and a *finite* width 2Δ. Let the midpoint of the interval 2Δ be positioned at a distance y from the x-axis. Then, according to (1) (with $\Delta\Omega$ omitted), the observed light intensity or the energy (per unit time) of all photons traveling *parallel* to the x-axis and coming through this aperture is given by

$$I_E(y) = \Delta z \int_{y-\Delta}^{y+\Delta} du \int_{-\infty}^{\infty} dx \varepsilon(\sqrt{u^2+x^2}) \equiv \int_{y-\Delta}^{y+\Delta} duI(u), \tag{2}$$

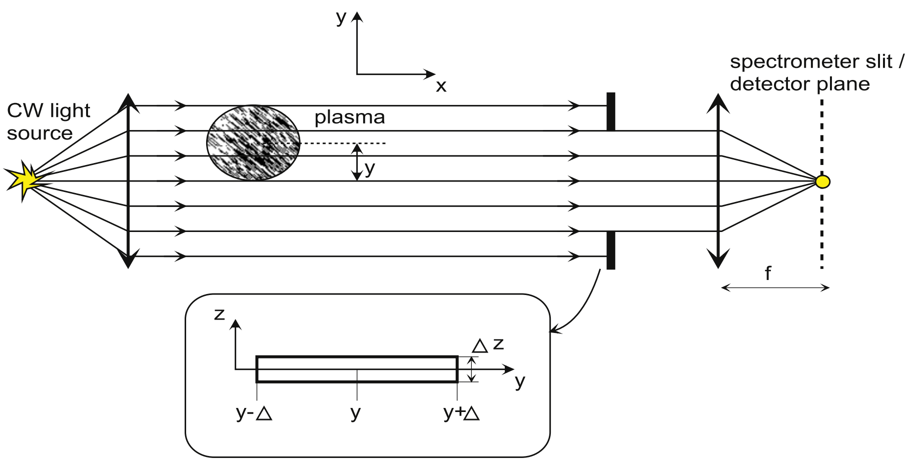

Fig. 1. An experimental setup for plasma absorption measurements with a large aperture. A CW light source is in focus of a lens to create a flux of parallel rays. A plasma plume with the symmetry axis normal to the figure plane is illuminated by the parallel rays. The parallel flux across a rectangular aperture is focused onto a detector (a spectrometer slit or CCD camera). The aperture plane is normal to the flux. It is positioned at a distance large enough to neglect contributions of the plasma plume emission. The aperture geometry is depicted in the inset at the bottom of the figure. The height Δz is small enough to neglect variations of the plasma absorption along the z-direction. The aperture width 2Δ can be of the size of the plasma plume or larger. The intensity in parallel rays is measured for different values of y, the distance between the plasma symmetry axis and the optical axis of the system. Measurements at a fixed value of y have no spatial resolution by themselves if Δ is large. The spatial resolution is *restored* by a numerical processing of data collected at different values of y.