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Review

What can we learn about laser-induced plasmas from Thomson scattering experiments

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ABSTRACT

This article describes laser Thomson scattering as applied to investigate laser-induced plasmas originating from gas breakdown or ablation of solid samples. Thomson scattering provides a reliable and direct mean of determining plasma electron density and electron temperature with high spatial and temporal resolution. Moreover, unlike e.g. optical emission spectroscopy, no assumptions about axial symmetry, thermodynamic conditions in the plasma or its chemical composition are needed to quantify these fundamental plasma parameters. Because Thomson scattering is inherently accompanied by Rayleigh light scattering, information about concentration of heavy particles and their temperature can be simultaneously derived from the experimental data. The heavy particle temperature and the electron one are the primary indicators of the plasma thermodynamic equilibrium.
The goals of this article are to describe the theory of Thomson scattering relevant for the studies of low-temperature laser-induced plasmas, discuss the instrumental details of Thomson scattering experiments, and review the results of studies in which this technique has been used to characterize laser-induced plasmas.

Contents

1. Introduction

Laser-induced plasmas (LIPs) have found numerous applications including pulsed laser deposition, precision laser micromachining, short wavelength sources for EUV lithography and last but not the least laser-induced breakdown spectroscopy (LIBS). In the last two decades LIBS has become a very popular analytical method due to many merits such as applicability to any kind of sample, no sample preparation, in-situ and remote sensing capabilities or real-time analysis. Recently, its analytical sensitivity has been largely improved by the application of double pulse [1] or resonant [2] laser configurations. However, from the analytical point of view, the quantitative issues of LIBS are still considered its Achilles heel. This is predominantly due to the complex nature of

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the laser–sample interaction and then due to the plasma–particle processes which are moreover strongly space and time dependent.

The quantitative elemental analysis by LIBS requires a thorough knowledge of atom, ion and electron number densities and their temperatures. These parameters are commonly deduced in indirect way from the optical emission spectra assuming plasma in local thermodynamic equilibrium (LTE) which is a basis for calibration-free LIBS [3]. The concept of LTE assumes balances between excitation and deexcitation processes as well as between ionization and recombination ones, all of them governed by collisions with electrons. Therefore, electron number density, electron temperature and their spatial and temporal distributions are crucial for better understanding of processes in LIP, reliable LTE validation and subsequent LIBS applications. Various aspects of LTE in plasmas are discussed in detail by van der Mullen [4] and recently recalled by Cristoforetti et al. [5] for the case of LIP.

For LIP, the electron number density n_e is usually determined from the Stark width of some reference emission lines [6] or much less often from the laser interferometric data [7]. However, the use of Stark widths is often difficult due to the limited number of spectral lines with Stark parameters of satisfactory accuracy with respect to both n_e and electron temperature T_e dependencies. On the other hand, the electron temperature of LIP is almost exclusively deduced from emission data applying either Boltzmann or Boltzmann–Saha equation or using the line-to-continuum intensity ratio [8]. The use of these methods for T_e determination is only possible and meaningful if the plasma is at least in partial local Saha equilibrium that is assuming the excitation and ionization temperatures equal to the electron temperature.

In general, optical emission spectroscopy (OES) is commonly used in LIP diagnostics because it has a relatively simple experimental arrangement and non-intrusive character. Nevertheless, OES does not allow for local measurements because the intensity is integrated along a line-of-sight. Local values of emission coefficients can be obtained only for layers either homogeneous or axially symmetric with the use of the Abel transformation. The major drawback of the Abel procedure is an error accumulation towards the axis of symmetry, i.e. towards the center of the LIP column where the great part of emission signal originates. Even slightly asymmetric plasma can result in large uncertainties of emission coefficients in central parts of LIP, hence in unreliable plasma diagnostics. The inhomogeneity of LIP with radial gradients of temperature can also result in strong reabsorption of the optical signals originating from the plasma core in its outer, much cooler zones. The effect of self-absorption must be corrected for by additional measurements with either a back mirror [9] or external light source configuration as it was performed by Karabourniotis et al. [10].

An alternative approach to OES in LIP studies is the application of active laser spectroscopy methods such as Raman, Rayleigh and Thomson scatterings. While Raman and Rayleigh scatterings consist in scattering by the bound electrons of atom, ion or molecule, Thomson scattering (TS) describes the interaction of photons with free charges present in plasmas (mainly with free electrons). Principal advantages of Thomson scattering 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, its equilibrium state or chemical composition.

TS is commonly used for the measurement of local electron and ion temperatures and electron number densities in a variety of laboratory and industrial plasmas [11–13]. Thomson scattering has became a standard tool in research of nuclear fusion plasmas where it is still the most reliable method for measurements of electron temperature. Over the last few decades a fast development of pulsed laser sources and intensified CCD cameras in particular, has made TS a popular method in investigations of industrial processing plasmas such as inductively coupled plasmas (ICP), microwave induced plasmas or plasma arcs and jets. All of them are classified as low temperature plasma sources with $T_e <$ 10 eV and n_e in the range 10^{15}–10^{23} m^{-3} resembling LIP characteristics.

The purpose of the current work is to discuss Thomson scattering method in application to low-temperature laser-induced plasmas of LIBS type. Despite great interest of research community in LIBS plasma and many difficulties encountered during its characterization by OES, the TS method has not been yet commonly employed. Some attempts to use laser light scattered from LIP in air were made by Diwakar and Hahn [14]. Although they claimed observation of significant TS signals, no quantitative analysis of the TS spectra was performed which would yield n_e and T_e. Recently, two groups, of Riley [15–19] and Dzierż ga [20–25], have successfully applied Thomson scattering method to study LIP plasmas originating from laser breakdown as well as from laser ablation. Some preliminary results were also reported by Liu et al. [26].

The structure of this paper is as follows. Section 2 gives brief description of the TS method and the way in which the electron temperature and density can be obtained from the scattered spectra. A special emphasis is placed on TS application for diagnostics of low-temperature and simultaneously high-density plasma but also on the related issue of plasma perturbation by the probe laser pulse. Section 3 contains a description of typical experimental setups used in TS measurements to characterize plasmas induced during laser gas breakdown and during laser ablation of solid samples. In Section 4, we present some results of TS experiments including electron density and temperature evolution and their spatial distributions, and studies of thermodynamic equilibria in LIP and of a propagating shockwave. Finally, Section 5 brings a short summary and conclusions.

2. Thomson scattering in brief

The scattered laser radiation from a plasma consists of dipole radiation by accelerating charged particles (Thomson scattering), dipole radiation due to polarization of neutrals and ions (both Rayleigh and Raman scatterings) and radiation due to polarization of dust particles (Rayleigh and Mie scatterings). The radiation scattered from free charges is mainly due to electrons because their mass is much lower than the mass of ions resulting in their much higher acceleration in an electric field of the laser and consequently large dipole radiation. Therefore, the most important information about plasma electrons is contained in TS spectra but many useful data about plasma can be also derived from the other scattering spectra.

In TS, as in any other laser scattering process, the frequency of the scattered photon is doubly Doppler shifted, since the radiating electron is moving relative to both the laser beam and the detector. As long as the motion of electrons in plasma is uncorrelated, the scattered partial waves add up in incoherent way and the resulting spectrum reflects the electron energy distribution. If this distribution is Maxwellian, electron temperature T_e can be derived by fitting the Gaussian profile to the TS spectrum. The electron number density n_e is then determined by calibrating the value of the total scattered power using either Rayleigh or Raman scattering spectra obtained for some reference gas [11].

However, in a plasma, electrons are not completely free but are exposed to the electric microfields produced by other charged particles. This weak coupling makes electrons undergo collective motion within a sphere with the size of the Debye shielding length $\lambda_D = (\varepsilon_0 k_B T_e / e^2 n_e)^{1/2}$ with ε_0 the permittivity of free space, k_B the Boltzmann constant and e the electron charge.

If the wavelength associated with the scattering wavevector $k = 4\pi \sin(\theta/2)/\lambda_L$ (see Fig. 1) is longer than the Debye length λ_D then the incident laser wave interacts with the shielded charges and the scattering is collective. In the opposite case, the scattering takes over a distance much smaller than λ_D and the laser wave interacts with individual electrons giving rise to the non-collective scattering.

The character (collective or non-collective) of TS is governed by the scattering parameter $\alpha \equiv 1/(k\lambda_D) \propto (\lambda_L/\sin(\theta/2))(n_e/T_e)^{1/2}$, introduced by Salpeter [27]. The scattering is non-collective when $\alpha \ll 1$ whereas collective TS proceeds for $\alpha \gg 1$.