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Local thermodynamic equilibrium in a laser-induced plasma evidenced by blackbody radiation

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Abstract

We show that the plasma produced by laser ablation of solid materials in specific conditions has an emission spectrum that is characterized by the saturation of the most intense spectral lines at the blackbody radiance. The blackbody temperature equals the excitation temperature of atoms and ions, proving directly and unambiguously a plasma in local thermodynamic equilibrium. The present investigations take benefit from the very rich and intense emission spectrum generated by ablation of a nickel-chromium-molybdenum alloy. This alternative and direct proof of the plasma equilibrium state re-opens the perspectives of quantitative material analyses via calibration-free laser-induced breakdown spectroscopy. Moreover, the unique properties of this laser-produced plasma promote its use as radiation standard for intensity calibration of spectroscopic instruments.

Keywords: Blackbody radiation; Local thermodynamic equilibrium; LIBS; Calibration-free.

1. Introduction

The size of laboratory plasmas being usually smaller than their characteristic length of radiation relaxation, the state of complete thermodynamic equilibrium is generally not established. Most of the generated photons escape from the plasma and only a minor fraction of them is reabsorbed. In atmospheric plasmas, the collision rates are large enough so that the characteristic lengths of particle relaxation are typically smaller than the plasma size [1]. This favors the establishment of local thermodynamic equilibrium: the plasma state can then be modeled in a simplified way using the statistical laws of equilibrium [2, 3]. However, as the characteristic times of particle relaxation and particle diffusion are similar, atmospheric density plasmas have nonuniform spatial distributions of temperature and densities [4, 5]. Plasmas produced by laser ablation of solids differ from other atmospheric plasmas [6]. According to their large initial density, the processes of relaxation are faster whereas the diffusion processes are slower [7, 8]. Due to the high initial pressure, the ablated material plume expands rapidly until it reaches a pressure equilibrium with the surrounding gas atmosphere [9, 10]. It was shown that for laser irradiation with ultraviolet nanosecond laser pulses, the plasma appears almost uniform if ablation occurs under an inert gas atmosphere [11]. The combination of both local thermodynamic equilibrium (LTE) and uniform spatial distribution qualifies such a plasma as an ideal radiation source that was evidenced by several signatures in the emission spectra of plumes produced by laser ablation of different materials [12].

In the present research note, we focus on a particular feature of such an ideal radiation source. Although the characteristic length of radiation relaxation is expected to exceed the plasma dimension for most wavelengths, the situation can be different for photons emitted by strong resonance lines. Their probability of being absorbed within the plasma is expressed by the optical thickness $\tau = \int \alpha(z)dz$, where α is the absorption coefficient and z the coordinate along the line of sight. In the case of LTE, the processes of absorption and emission are related through Kirchhoff's law of thermal radiation $\varepsilon_\lambda/\alpha = U_\lambda$. Here, ε_λ is the spectral emission coefficient and U_λ is the blackbody spectral radiance given by Planck's law. For a uniform spatial distribution, the spectral radiance of the plasma in LTE is given by [13]

$$B_\lambda = U_\lambda(1 - e^{-\tau}). \tag{1}$$

Here $\tau = \alpha L$, where L is the plasma size along the line of sight.

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