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Abstract

We investigated the time evolution of pressure in the plume generated by laser ablation with ultraviolet nanosecond laser pulses in a near-atmospheric argon atmosphere. These conditions were previously identified to produce a plasma of properties that facilitate accurate spectroscopic diagnostics. Using steel as sample material, the present investigations benefit from the large number of reliable spectroscopic data available for iron. Recording time-resolved emission spectra with an echelle spectrometer, we were able to perform accurate measurements of electron density and temperature over a time interval from 200 ns to 12 μs. Assuming local thermodynamic equilibrium, we computed the plasma composition within the ablated vapor material and the corresponding kinetic pressure. The time evolution of plume pressure is shown to reach a minimum value below the pressure of the background gas. This indicates that the process of vapor-gas interdiffusion has a negligible influence on the plume expansion dynamics in the considered timescale. Moreover, the results promote the plasma pressure as a control parameter in calibration-free laser-induced breakdown spectroscopy.

Keywords: Kinetic pressure; Particle diffusion; Plasma modeling; Spectra simulation; Calibration-free LIBS.

1. Introduction

In pulsed laser ablation under conditions typically applied during material analyses via laser-induced breakdown spectroscopy (LIBS), the plasma generated at the sample surface is characterized by a temperature of several eV and a density close to solid density during the initial stage of expansion [1]. The plume undergoes a rapid explosion-like expansion until it reaches a pressure equilibrium with the surrounding compressed gas. On a longer timescale, the processes of heat and particle diffusion take place and contribute to the further evolution of plasma properties [2]. The precise scenario of the LIBS plasma development depends naturally on the laser irradiation conditions and the gas environment. Compared to short pulse laser excitation, nanosecond laser ablation takes advantage from the interaction of the laser beam with the vaporized material. This process changes the thermodynamic pathway of the laser-heated matter and leads to larger degrees of atomization and excitation [3]. In the case of ultraviolet (UV) radiation, laser heating occurs exclusively in a tiny volume close to the sample surface where the plasma density is high enough. This leads to an increased laser energy coupling towards the sample for the short-wavelength radiation. Plasma screening via laser energy absorption by the surrounding atmosphere is negligible and the background gas is heated by the shock wave and the interaction with the ablated vapor plume only [4].

The background gas has a strong influence on the properties of the laser-produced vapor plasma. In a molecular gas, such as ambient air, inelastic collisions play a significant role in the vapor-gas interaction. The energy loss at the vapor-gas contact front leads to the formation of a cold peripheral layer within the vaporized material [5]. The situation is different for ablation in an inert gas. Because of the large excitation energy, the plasma electrons interact with the background gas atoms mainly through elastic collisions, conserving thus their energy. This suppresses the formation of the cold border within the ablated vapor plume. The plasma appears uniform and diagnostics are easy to handle as they do not require space-resolved measurements. If furthermore the assumption of local thermodynamic equilibrium (LTE) is valid, the laser-produced plasma

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