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Ablation plume structure and dynamics in ambient gas observed by laser-induced fluorescence imaging spectroscopy

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ABSTRACT

The dynamic behavior of an ablation plume in ambient gas has been investigated by laser-induced fluorescence imaging spectroscopy. The second harmonic beam from an Nd:YAG laser (0.5–6 J/cm^2) was focused on a sintered oxide pellet or a metal chip of gadolinium. The produced plume was subsequently intersected with a sheet-shaped UV beam from a dye laser so that time-resolved fluorescence images were acquired with an intensified CCD camera at various delay times. The obtained cross-sectional images of the plume indicate that the ablated ground state atoms and ions of gadolinium accumulate in a hemispherical contact layer between the plume and the ambient gas, and a cavity containing a smaller density of ablated species is formed near the center of the plume. At earlier expansion stage, another luminous component also expands in the cavity so that it coalesces into the hemispherical layer. The splitting and coalescence for atomic plume occur later than those for ionic plume. Furthermore, the hemispherical layer of neutral atoms appears later than that of ions; however, the locations of the layers are nearly identical. This coincidence of the appearance locations of the layers strongly suggests that the neutral atoms in the hemispherical layer are produced as a consequence of three-body recombination of ions through collisions with gas atoms. The obtained knowledge regarding plume expansion dynamics and detailed plume structure is useful for optimizing the experimental conditions for ablation-based spectroscopic analysis.

1. Introduction

In the past few decades, laser ablation has become a key technology having extensive practical and research-oriented applications including pulsed laser deposition (PLD) of thin films [1], ultraviolet (UV) light source for lithography [2], laser-induced breakdown spectroscopy (LIBS) [3], laser-ablation inductively coupled plasma mass spectrometry [4], and fundamental atomic spectroscopy [5], where the ablation plume is produced under various experimental conditions. Accompanying this expansion of the fields of laser ablation application, a significant amount of research on plume characteristics has been conducted mainly by means of laser-induced plasma emission spectroscopy [6]. In emission spectroscopy, a majority of the deduced information pertains to highly excited atoms in a plume. However, for some applications, evaluating the behavior of ground state and metastable atoms (hereafter shortly ground state atoms) in detail is desirable. For instance, many researchers are currently developing LIBS techniques with improved sensitivity utilizing both ground state and highly excited atoms, such as double pulse LIBS [7,8], combined LIBS and laser-induced fluorescence (LIF) [9,10], resonant LIBS [11], resonance-enhanced LIBS [12], spark discharge LIBS [13], and microwave-assisted LIBS [14–16]. Additionally, the properties of PLD-produced thin films, such as thickness and stoichiometry, are determined to a great extent by the spatial distribution of ground state atoms because this is a major component of the ablated species in a plume.

Presently, we have investigated various characteristics of ground state atoms in ablation plumes using laser ablation absorption spectroscopy (LAAS) [17–19] to develop remote isotope analysis for highly radioactive nuclear fuels [20–22]. We used a low pressure gas environment because the measurement of highly resolved absorption spectra for ablated species requires a reduction of Stark and pressure broadenings. Because the plume expansion in this case is considerably affected by the ambient gas, it is of great importance to evaluate the spatiotemporal distribution of the ablated species in gas for sensitive analysis. Our previous study revealed that the absorption spectrum for ablated species splits into two symmetrical peaks under particular experimental conditions, and the splitting interval gradually decreases with time after ablation [20]. From the splitting behavior owing to the Doppler effect, it was inferred that most of the ablated species accumulate in a hemispherical layer at a boundary between the expanding plume and the ambient gas, and that a cavity containing a lower density of ablated species develops in the region surrounded by the layer. Because very few studies have reported such a hollow plume structure, experimental confirmation of the expected distribution and a detailed analysis of the formation of this structure are desirable for applications using the laser ablation technique.

Most previous studies concerning plume structure have utilized emission imaging spectroscopy [23–28]. From the observed emissivity

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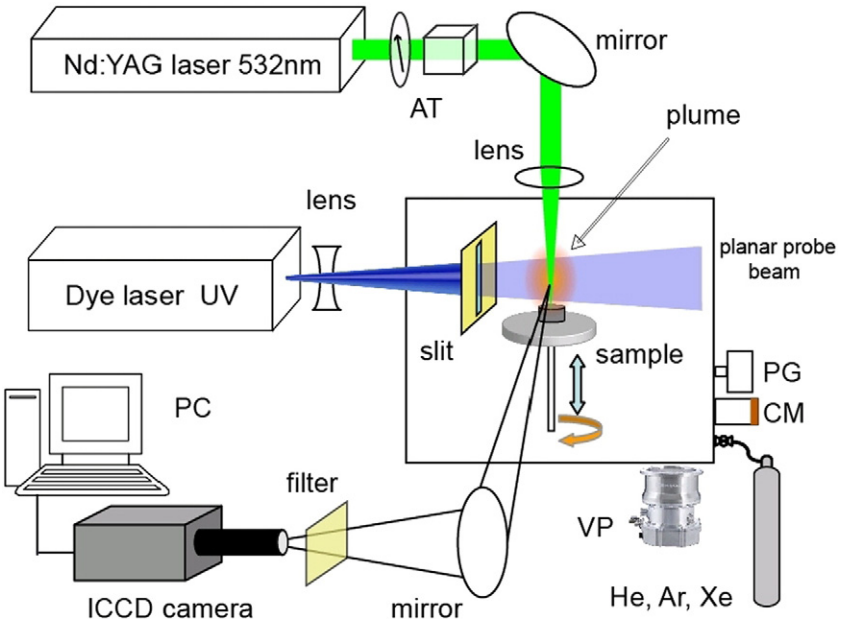

Fig. 1. Experimental setup for LIF imaging spectroscopy. VP, PG, CM and AT mean vacuum pump, convection-enhanced Pirani gauge, capacitance manometer and attenuator consisting of polarizing beam-splitter cube and half-wave plate. LIF images of ablation plume were acquired with an ICCD camera from an orthogonal direction to the planar probe beam.

distributions, several researchers have estimated the cross-sectional number density distributions of highly excited atoms using Abel inversion [29,30]. In contrast, for the study of the distribution of ground state atoms in a plume, LIF and absorption imaging techniques have been employed. For example, Gilgenbach et al. demonstrated dye-laser resonance absorption photography in the early 1990s [31]. Around the same

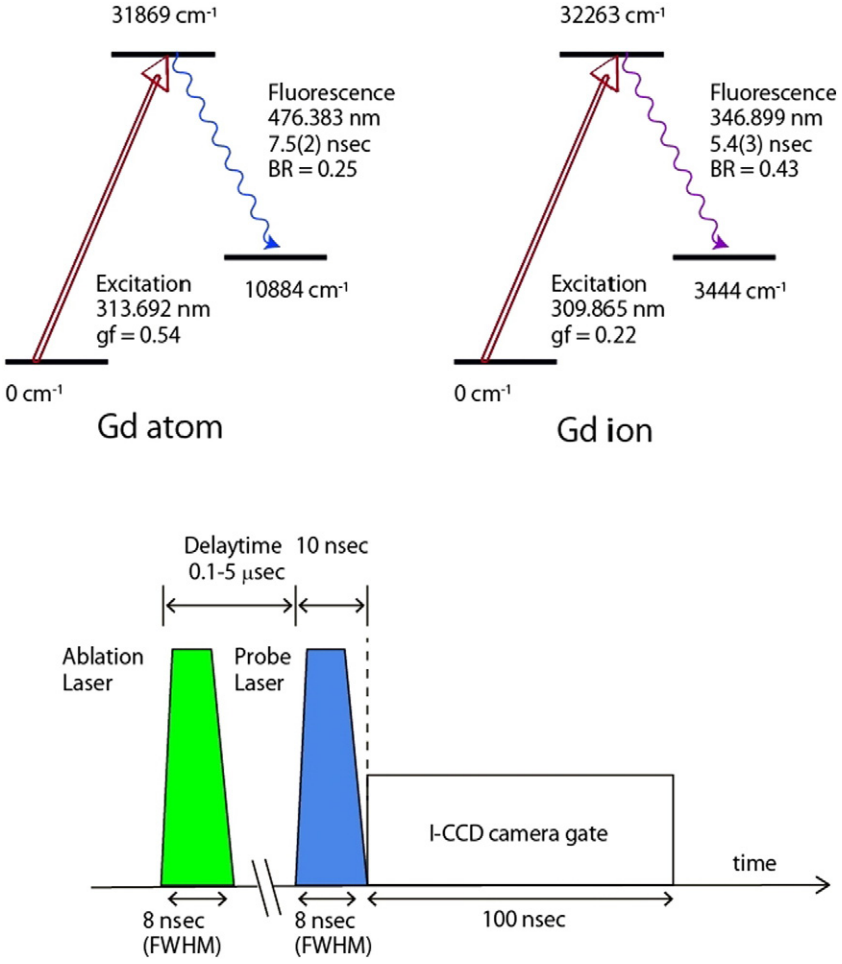

Fig. 2. Excitation and fluorescence transition schemes of Gd (upper), and laser firing and camera gate timings (lower).