Spectrochimica Acta Part B 66 (2011) 57–66

Contents lists available at ScienceDirect

Spectrochimica Acta Part B

journal homepage: www.elsevier.com/locate/sab

Time-resolved optical emission spectroscopic measurements of He plasma induced by a high-power CO_2 pulsed laser

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ARTICLE INFO

Article history:
Received 15 March 2010
Accepted 2 December 2010
Available online 10 December 2010

Keywords:
Laser-induced breakdown spectroscopy (LIBS)
Time-resolved optical emission spectroscopy
High-power IR CO_2 pulsed laser
Velocity and kinetic energy distributions
Time-of-flight measurement
Plasma diagnostics
Helium

ABSTRACT

Laser-induced breakdown spectroscopy of helium plasma, initially at room temperature and pressures ranging from 12 to 101 kPa was investigated using a transverse excitation atmospheric CO_2 pulsed laser ($\lambda = 9.621$ and 10.591 μm, a full width at half maximum of 64 ns, and an intensity from 1.5 to 5.36 GW cm^{-2}). The helium breakdown spectrum is mainly due to electronic relaxation of excited He, He^+ and H. Plasma characteristics were examined in detail on the emission lines of He and He^+ by the time-integrated and time-resolved optical emission spectroscopy technique. Optical breakdown threshold intensities, ionization degree and plasma temperatures were obtained. An auxiliary metal mesh target was used to analyze the temporal evolution of the species in the plasma. The results show a faster decay of the continuum emission and He^+ species than in the case of neutral He atoms. The velocity and kinetic energy distributions for He and He^+ species were obtained from time-of-flight measurements. Electron density in the laser-induced plasma was estimated from the analysis of spectral data at various times from the laser pulse incidence. Temporal evolution of electron density has been used for the estimation of the three-body electron–ion recombination rate constant.

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1. Introduction

The development of high-power lasers has intensified the discovery and investigation of many new phenomena induced in a gas by laser radiation, and the interaction of the latter with ionized gases and plasmas [1]. Irradiation of a gas with a high-power pulsed laser can result in an explosive emission of atoms, molecules, ions and electrons from dielectrical breakdown [2,3]. The interest in laser-induced breakdown (LIB) is due to a number of potential applications of high electron density and high-temperature plasmas [4–6]. Because of the transient characteristics of the plasma plume created by LIB, the optical emission spectroscopy (OES) technique with time and space resolution is especially appropriate to obtain information about the behavior of the formed species as well as to study the dynamics of the plasma expansion. The temporally-resolved LIB technique in He has been used by several researchers [7–13] using high-power lasers. The emission spectra of LIB plasma produced by a CO_2 laser in different gases have been reported recently by us [14–16]. For He, it has been demonstrated by time-integrated and time-resolved OES [17] that a TEA-CO_2 laser with the help of a metal sub-target generates helium breakdown plasma in which the helium meta-stable excited state is produced.

In this paper, we present a study of the helium plasma (He initially at room temperature and pressures ranging from 12 to 101 kPa) directly produced by a high-power IR CO_2 pulsed laser ($\lambda = 9.621$ and 10.591 μm; a full width at half maximum (FWHM) of 64 ns; and power densities ranging from 1.5 to 5.36 GW cm^{-2}). Only He and its impurities are present in the samples; the metal sub-target is only used for fixing the temporal initiation of the breakdown. We discuss thermo-chemical processes induced by the laser pulse on helium and we evaluate plasma changes, which are of fundamental importance in establishing the mechanisms responsible for the plasma emission. The emission observed in the plasma region is mainly due to electronic relaxation of excited He, H (as an impurity) and He^+ ionic fragments. The characteristics of the spectral emission intensities from different species have been investigated as functions of the He pressure and laser irradiance. Optical breakdown threshold intensities in helium at 10.591 μm have been measured. In addition, we also present new results obtained from the time-resolved spectroscopic analysis of the LIB He plasma. We discuss the dynamics of the plasma expansion and formation of He and He^+ species for different delay times with respect to the beginning of the laser pulse. The velocity and kinetic distributions for He and He^+ are obtained from time-of-flight (TOF) measurements using time-resolved OES. Possible mechanisms for the production of these distributions are discussed. Intensities of some lines from He and He^+ were used for determining electron temperature and their Stark-broadened profiles were employed to calculate electron density. The temporal behavior of the electron

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doi:10.1016/j.sab.2010.12.001

number density has been employed for the estimation of three-body recombination rate constant and recombination time.

2. Experimental procedure

The experimental arrangement used in this research is shown in Fig. 1. The experiments were carried out with transverse excitation atmospheric (TEA) CO_2 laser (Lumonics model K-103) operating on an 8:8:84 mixture of CO_2:N_2:He, respectively. The laser delivered up to 3160 mJ at a wavelength of 10.591 μm, leading to an estimated power of 49.5 MW, an intensity (power density or irradiance) of 6.31 GW cm^{-2}, a fluence of 402 J cm^{-2}, a photon flux of 3.4×10^{29} photon cm^{-2} s^{-1}, and an electric field of 1.63 MV cm^{-1} on the focal position. The focused-spot area (7.85×10^{-3} cm^2) of the laser beam was measured with a pyroelectric array detector (Delta Development Mark IV). The temporal shape of the TEA-CO_2 laser pulse consisted of a prominent spike of ≈64 ns (FWHM) followed by a long lasting tail of lower energy and about 3 μs duration. A beam splitter was used to redirect 10% of the laser pulse energy on a pyroelectric detector (Lumonics 20D) or on a photon-drag detector (Rofin Sinar 7415) for energy or temporal shape monitoring and triggering, respectively, through a digital oscilloscope (Tektronix TDS 540). The primary laser beam was angularly defined and attenuated by a diaphragm of 17.5 mm diameter before entering to the He cell. The laser pulse energy was varied with the aid of several calibrated CaF_2 attenuating plates. The shot-to-shot fluctuation of the laser energy was approximately 5%. The pulsed laser light from the CO_2 laser was focused by two NaCl lenses of 24 or 40 cm focal distance. The primary helium studied in these experiments was of high purity >99.995% (<5 ppm H_2O, <20 ppm N_2, <5 ppm O_2 and <1 ppm C_nH_m). In order to fix the temporal and spatial origin, a metal mesh (0.7 mm × 0.7 mm stainless steel) was used as the auxiliary target. Although the metal mesh itself can be ablated, in our experimental conditions, mainly time- and space-resolved emission spectra from the helium plasma were recorded. The light emitted from the He plasma was imaged 1:1 by a quartz lens (focal length 4 cm, f-number = f/2.3) onto the entrance slit of the spectrometer. Two spectrometers were used: 1/8 m Oriel spectrometer (25 μm slit and a grating of 1200 grooves mm^{-1}) at a resolution of ≈0.13 nm in first-order and a 0.32 m ISA Jobin Yvon Spex (Model HR320) (adjustable slit and a holographic grating of 2400 grooves mm^{-1}) at a resolution of ≈0.02 nm in first-order. The distance between the He plasma axis and entrance slit was $y = 16$ cm. For time-resolved measurements, optical emission accompanying the laser-induced He plasma was viewed in a XZ parallel plane to the front face of the metal mesh for a fixed distance of $z = 5$ mm along the plasma Y axis. The spectra were recorded by a gateable ICCD (Andor iStar DH-734). The spectral window for each experiment with this detection system was about 12 nm. For synchronization, the CO_2 laser was operated at the internal trigger mode and the ICCD detector was operated in external and gate modes. The external trigger signal generated by the laser was fed through the scope and delay generator into the back of the ICCD detector head. The total insertion delay (45 ± 2 ns) is the total length of time taken for the external trigger pulse to travel through the digital delay generator and gate so that the ICCD will switch on. The time jitter between the laser and the fast ICCD detector gate was about ± 2 ns. The delay time t_d is the time interval between the arrival of the laser pulse on the metal mesh and the activation of the ICCD detector. The gate width time t_w is the time interval during which the plasma emission is monitored by the ICCD. Both parameters were adjusted by the digital delay generator of the ICCD detector. The CO_2 laser pulse picked up with the photon-drag detector triggered a pulse generator (Stanford DG 535) through the scope and this pulse was used as the external trigger in the ICCD camera. The laser pulse and the gate monitor output were displayed in a digital oscilloscope. In this way, by using the output of the photon-drag detector, the oscilloscope, the delay pulse generator and the gate monitor output of the ICCD camera, the gate width time t_w and the delay time t_d could be adjusted without insertion time. Several (Cu/Ne, Fe/Ne and Cr/Ar) hollow cathode lamps (HCL) were used for the spectral wavelength calibration of the spectrometers. The

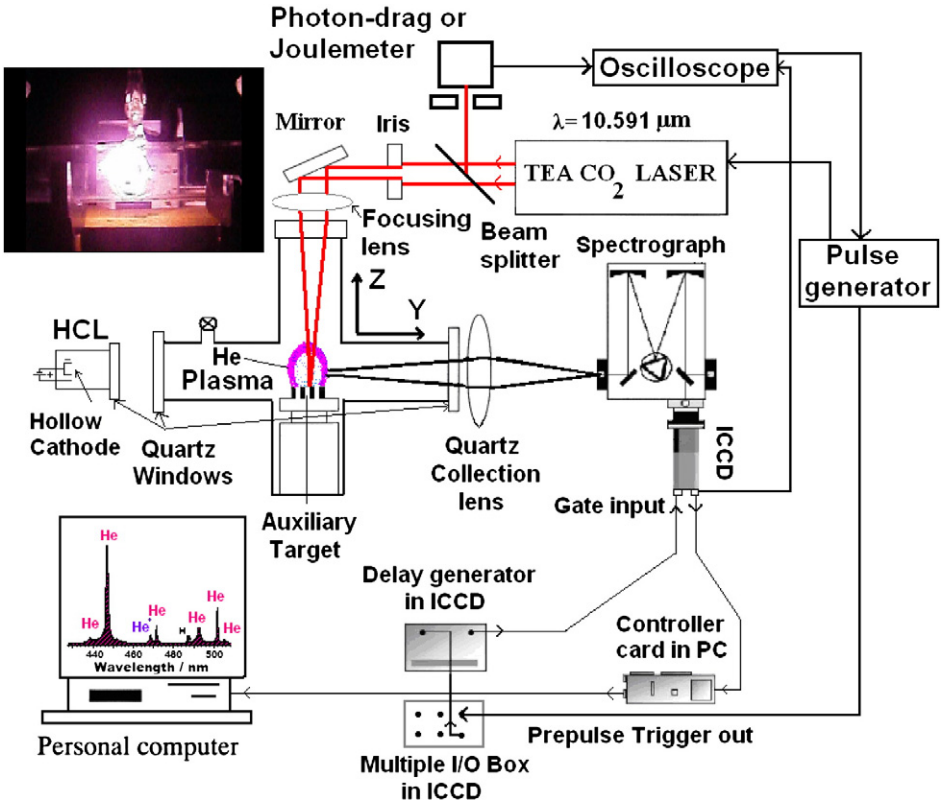

Fig. 1. Schematic diagram of the experimental setup of the time gated ICCD for TEA-CO_2 pulsed laser He breakdown diagnostics.

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