Temporal characterization of CO_2 plasma induced by a high-power TEA-CO_2 pulsed laser

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It is well known that for sufficient high electric field strengths in an insulating medium as CO_2 a breakdown or plasma can occur. Breakdown in gases can be initiated by pulsed high-power lasers. This work reports studies on laser-induced breakdown (LIB) spectroscopy in CO_2, initially at room temperature and pressures ranging from 6.4 to 101 kPa, by using a high-power transverse excitation atmospheric (TEA) IR CO_2 pulsed laser (λ=10.532 μm, 64 ns (full width at half maximum), 1.2-5.6 GW×cm^{-2}). Because of the transient characteristics of the plasma created by the laser, optical emission spectroscopy (OES) technique with time and space resolution is especially appropriate to obtain information about the behaviour of the formed species as well as to study the dynamics of the plasma expansion. We discuss the dynamics of the plasma expansion and formation of different atomic, ionic, and molecular species for different delay times with respect to the beginning of the laser pulse. Detailed temporal studies of LIB on carbon and air have been recently carried out in our laboratory [1].

The experiments were carried out with a high-power TEA-CO_2 laser (Lumonics model K-103). The temporal shape of the TEA-CO_2 laser pulse consisted in a prominent spike of a full width at half maximum $\approx$ 64 ns 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 or on a photon-drag detector for energy or temporal shape monitoring and triggering, respectively, through a digital oscilloscope. The laser-pulse energy was varied with the aid of several calibrated CaF_2 attenuating plates. The pulsed laser light from the CO_2 laser was focused by a NaCl lenses. Optical emission from the CO_2 plasma was collected onto the entrance slit of different spectrometers. The spectra were recorded by a gateable ICCD (Andor iStar DH-734). The temporal history of the plasma was obtained by recording the emission at predetermined delays and at a fixed gate width..

The strong emission of the CO_2 plasma, at medium-vacuum conditions (4 Pa), shows ionized excited species (O^+, C^+, and C^{2+}) and neutral O and C atoms. The medium-weak emission is due to electronic relaxation of excited species O^{2+}, C^{3+}, N^+, N, H and molecular band systems of $C_2(d^3\Pi_g\text{-}a^3\Pi_u;\ e^3\Pi_g\text{-}a^3\Pi_u;\ E^1\Sigma_g^+\text{-}A^1\Pi_u;\ A^1\Pi_u\text{-}X^1\Sigma_g^+)$, $CN(B^2\Sigma^+\text{-}X^2\Sigma^+;\ A^2\Pi\text{-}X^2\Sigma^+)$, $O_2(b^1\Sigma_g^+\text{-}X^3\Sigma_g^-)$, $N_2(C^3\Pi_u\text{-}B^3\Pi_g)$ and $N_2^+(B^2\Sigma_u^+\text{-}X^2\Sigma_g^+)$. Figure 1 displays an overview of the OES of the LIB in carbon dioxide compared with the atomic/ionic lines of C, C^+, C^{2+}, C^{3+}, O, O^+, O^{2+}, N and N^+. In this experiment the laser delivered 1323 mJ at a wavelength of 10.532 μm, leading to an estimated power of 20.7 MW, intensity (power density or irradiance) of 2.64 GW×cm^{-2}, fluence of 169 J cm^{-2}, photon flux of 1.3×10^{29} photon×cm^{-2}×s^{-1}, and electric field of 1.0 MV×cm^{-1} on the focal position.

The assignments of atomic/ionic lines of carbon, oxygen and nitrogen are indicated by stick labels. In the lower part of this figure, we indicated in a column graph the relative intensities of the observed C, C^+, C^{2+}, O, O^+, O^{2+}, N and N^+ lines listed in the NIST Atomic Spectral Database. There is a good agreement between the line intensities tabulated in NIST and the measured intensities observed. Excitation temperatures and electron number densities were estimated by means of C^+ and O^+ lines. The characteristics of the spectral emission intensities from different species have been investigated as function of the CO_2 pressure and laser irradiance.

Experimental threshold power densities for CO_2 is measured for the TEA-CO_2 laser at λ=10.532 μm. The minimum laser intensity required to form plasma is called the breakdown threshold. The threshold power density is dependent on the kind of laser, laser wavelength, pulse width, beam size of the focal volume, and gas pressure. The pressure dependence of the laser-induced breakdown thresholds is not in agreement with multiphoton ionization (MPI) process which predicts very weak pressure dependence for the threshold power density, while it is in qualitative agreement with electron cascade ionization. A minimum in the variation of the threshold power density versus pressure is predicted by the classical theory. In our experiments, a minimum in the threshold power density versus pressure curve is observed. Therefore, starting from our experimental observations and calculations, we can conclude that although, the first electrons must appear via MPI or natural ionization, electron impact is the main mechanism responsible for the breakdown in carbon dioxide.

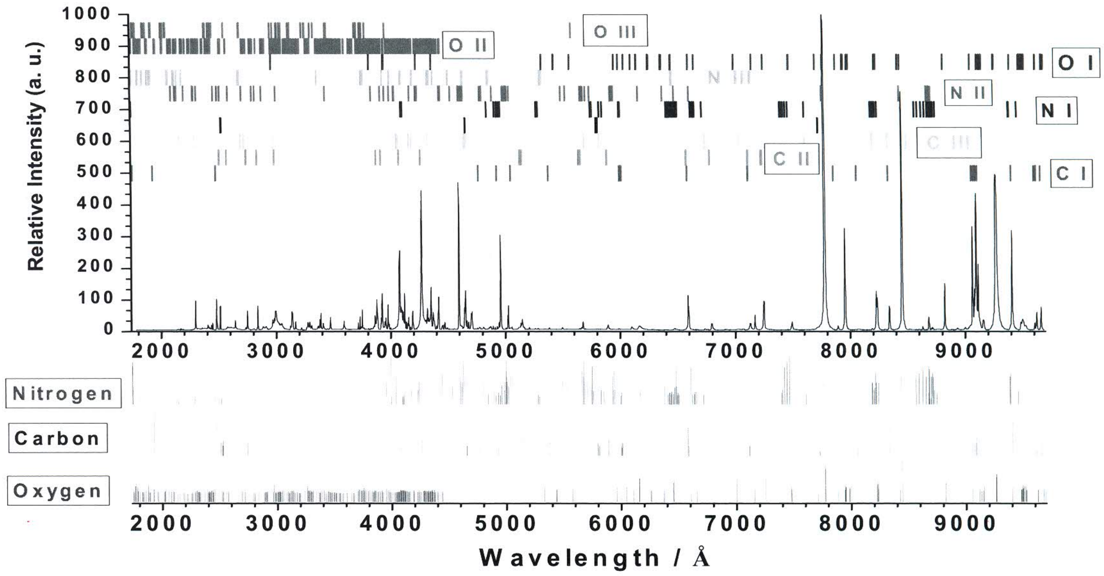

Fig. 1. An overview of the LIB emission spectrum of carbon dioxide (33 kPa) plasma induced by a TEA-CO_2 high-power pulsed laser (2.6 GW cm^{-2}) and assignment of some atomic/ionic lines.

The structure of the LIB plasma in CO_2 or in other gases is complex, and indeed there may be several isolated plasma regions with near ellipsoidal shape, produced along the laser axis. These multiple collinear plasmas difficult the definition of the focal region. Focusing the laser on a metal mesh, allow us to fix the temporal and spatial origin. Time-resolved OES is used to analyze mesh-initiated CO_2 breakdown plasma. The results show a faster decay of the continuum emission and ionic species than in the case of neutral atoms. The velocity and kinetic energy distributions for different species are obtained from time-of-flight measurements. Electron density in the laser-induced plasma was estimated from the Stark broadening method by analyzing different spectral lines at various times from the CO_2 laser pulse incidence. Temporal evolution of electron density has been used for the estimation of the three-body electron-ion recombination rate constant and recombination time.

1. J.J. Camacho, L. Diaz, M. Santos, L.J. Juan and J.M.L. Poyato, *J. Appl. Phys.* **106**, 033306 (2009); *J. Appl. Phys.* **107**, 083306 (2010).

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