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In this contribution we present Two-photon Absorption Laser Induced Fluorescence (TALIF) measurements on expanding plasmas produced by a cascaded arc in Ar/H_2 and Ar/N_2 mixtures. From these measurements axial and radial profiles for the density, the temperature, and the radial velocity of the atomic species are obtained.

Introduction

When a cascaded arc created plasma is expanded into a vacuum vessel, a versatile high quality particle beam is obtained that has been shown to have very promising features [1,2]. An important application of such an expanding plasma is its use for fast deposition of e.g. amorphous silicon (a-Si:H) or carbon (a-C:H) [2,3]. For these applications, but also from a fundamental point of view, it is interesting to study the composition of the beam, i.e. the degree of dissociation, excitation, and ionization, as a function of externally controlled parameters, such as flow, pressure, arc current, etc.
Several laser-based diagnostic techniques, like Thomson/Rayleigh scattering, CARS, and LIF are used to probe the local particle densities in the expanding plasma and their energy distribution. These techniques have the general advantage to be non-intrusive and very sensitive, while their signal is not disturbed by locally varying conditions. However, species-selective monitoring of atoms in their ground-state requires the use of high energetic photons because of the large energy spacings involved. Experimentally demanding techniques that are necessary for single-photon excitation can be overcome when multi-photon excitation is applied.

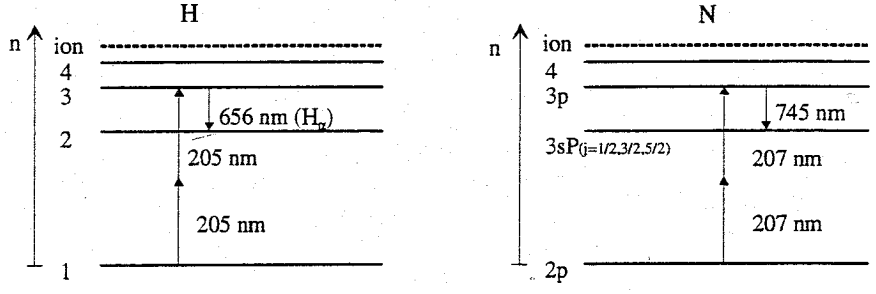

Fig. 1: Energy level schemes for the two-photon absorption laser-induced fluorescence (TALIF) measurements on H (left) and N (right).

In this contribution we present measurements on atomic H and N using two-photon excitation and detection of the resulting non-resonant laser-induced fluorescence (LIF). The corresponding energy level schemes are shown in Fig. 1.
Atomic H is excited with two 205.14 nm photons from the 1s ^2S ground state to the 3d ^2D and 3s ^2S states. The excitation is monitored by detection of the fluorescence on the Balmer-α line at 656.3 nm. Atomic N is excited from the 2p $^4S^0$ ground state to the 3p $^4S^0$ state with two 206.7 nm photons. The resulting fluorescence to the 3s ^4P states is detected around 745 nm.

Experimental

A scheme of the experimental set-up is depicted in Fig. 2. The cascaded arc plasma source has already been described in detail elsewhere [4]. Here it is operated on a 40 A dc current. The plasma expands from a 4 mm diameter channel into a roots-blower pumped vessel with a backing pressure of 10 Pa. For the measurements on H an 6:1 Ar/H_2 mixture is used with 3.0 slm Ar-flow and 0.5 slm H_2-flow. The plasma conditions for this case are close to those used for deposition of a-C:H and a-Si:H. For the measurements on N a nearly 100% N_2-flow (2.0 slm) is used with only a small fraction of Ar (0.05 slm) to stabilize the burning plasma. The cascaded arc plasma source is mounted on a translation arm. Spatial scans through the expanding plasma can be made by moving the cascaded arc relative to the laser beam and detection optics.

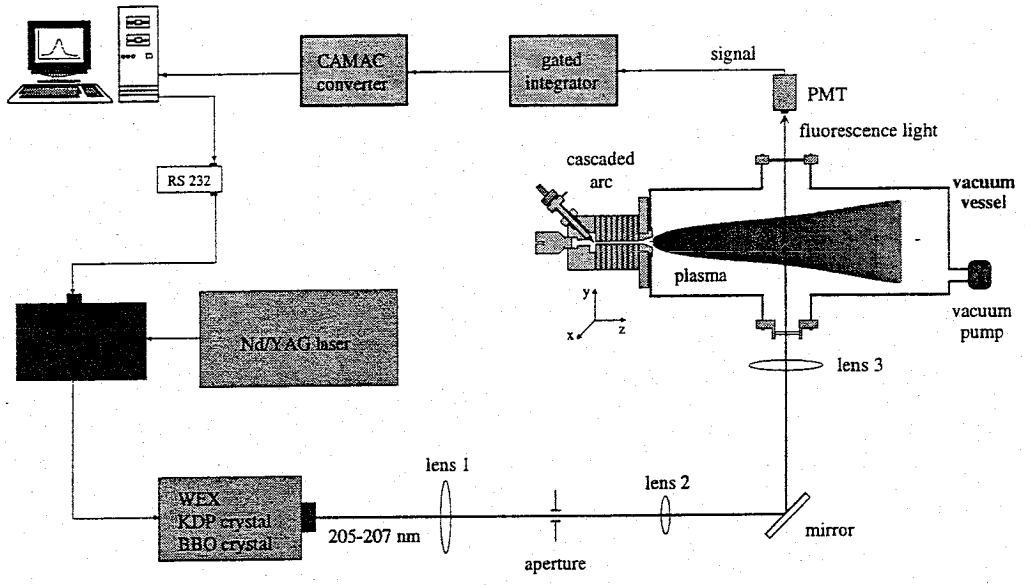

Fig. 2: Scheme of the experimental set-up. The cascaded arc plasma source is mounted on a translation arm that allows for spatially resolved measurements.

The laser system that is used to produce the tunable UV-radiation is based on a pulsed Nd:YAG/dye-laser combination. The frequency-doubled output (250 mJ pulse energy) of a 50 Hz injection-seeded Nd:YAG-laser (Spectra-Physics, GCR-230) is used to pump a tunable dye-laser (Spectra-Physics, PDL-3). The dye-laser has a bandwidth of 0.07 cm^{-1} and is used around 615 nm for the measurements on H and around 618 nm for the N measurements. The output of the dye-laser (typically 60 mJ) is frequency-tripled in two stages using non-linear optical techiques. In the first stage the light is frequency-doubled in a KD*P-crystal. In the second stage this frequency-doubled UV light is mixed with the residual red dye-laser output in a BBO-crystal where typically 0.5 mJ of tunable UV light around 205 nm with an estimated bandwidth of 0.2 cm^{-1} is produced via sum-frequency generation.
The frequency-tripled laser light is imaged via a lens-pinhole-lens combination onto a 60 cm lens that focusses the light into the plasma. The laser-induced fluorescence originating from the focus is imaged with a spatial resolution of 2 mm, in a direction perpendicularly to the laser beam, onto a photo-multiplier (Hamamatsu, R928). The continuous background light emitted by the plasma is strongly reduced by an optical filter in front of the photo-multiplier tube (PMT). In case of the H-fluorescence a H_α bandpass filter is used, whereas in the case of N-fluorescence a highpass filter is used that is transparant above 730 nm. The background signal is further reduced by accumulating the PMT-signal with a gated integrator only during a preset time interval of some 200 ns after the arrival of the laser pulse. The resulting signal is digitized by a CAMAC converter, and then read into a PC that also controls the wavelength scanning of the dye-laser.

Results

All fluorescence measurements are obtained by carefully tuning the excitation laser and recording the resulting spectral profile of the two-photon absorption for various axial and radial positions. The spectral scans are all fitted to a Gaussian profile. From these fits three local parameters can be obtained in principle. The local density is obtained from the integrated intensity (i.e. the area under the curve), while the local temperature is derived from the Doppler-width of the spectral profile. In addition, the radial component of the velocity can be determined from the absolute shift of the center frequency of the spectral profile. In order to obtain quantitative velocity data, the wavelength of the laser needs to be calibrated.

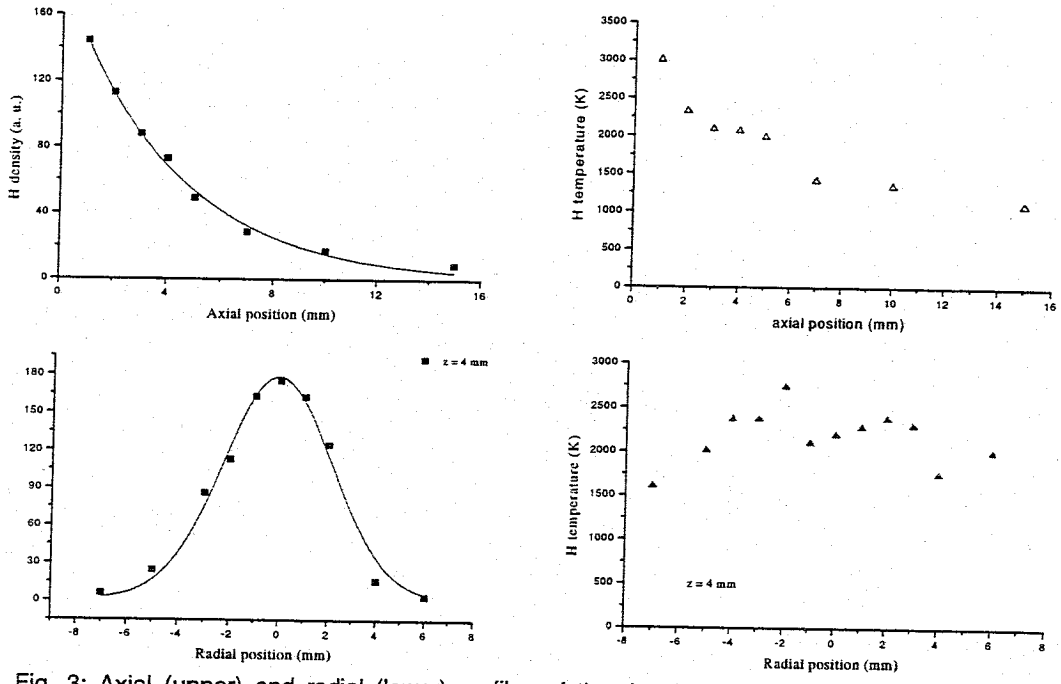

Fig. 3: Axial (upper) and radial (lower) profiles of the density (left) and temperature (right) of atomic H. The radial scans are taken at z =4 mm.

The axial and radial profiles of the density and temperature of atomic H are shown in Fig. 3. The axial density profile can be fitted well to an exponential decay with a 1/e length of 4 mm (solid line). The temperature on the axis decreases from about 3000 K near the nozzle to 1000 K at a distance of 16 mm. Radial scans show for the density a Gaussian-like shape with a clear maximum in the centre and for the temperature a plateau in the centre with the temperature falling down at the edges. The axial line density, that is the density integrated over one radial plane, appears to be almost constant. Assuming that the axial velocity is constant, this implies that the total amount of atomic hydrogen is conserved during the expansion.

For atomic N the axial and radial profiles of the density and temperature are shown in Fig. 4. Both the axial density and temperature profile show evidence of a shock region around 100 mm from the nozzle. The radial density profiles are again well approximated by a Gaussian, while the radial temperature profile is less well-behaved.

Discussion and outlook

The TALIF technique enables spatially resolved measurements of the density, the temperature, and the velocity of atoms in e.g. expanding plasma's. In our experimental set-up an absolute wavelength calibration will immediately yield absolute values for the radial velocity of atomic H and N. A

calibration based on the simultaneous measurement of the absorption spectrum of I_2 is currently being implemented. In addition, we plan to measure the axial velocities directly by applying the excitation laser anti-parallel or under a small angle with the symmetry axis of the expansion.

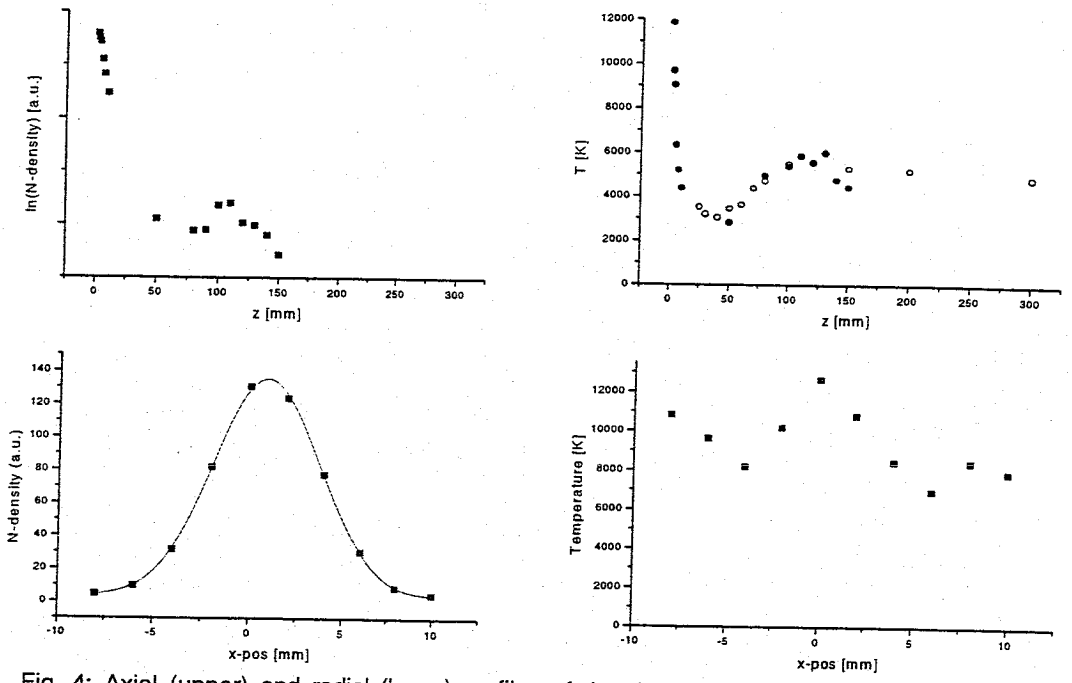

Fig. 4: Axial (upper) and radial (lower) profiles of the density (left) and temperature (right) of atomic N. The radial scans are taken at z =5 mm.

The density profiles measured via TALIF are relative and need to be calibrated in order to obtain absolute values for the number densities. For this the measurements need to be correlated to the TALIF-signal of an independent source that produces a well-known amount of atomic H and/or N. This can be done using a titration reaction in a flow tube reactor [5]. Atomic H can be produced in a flow tube reactor by dissociation of molecular H_2 in a microwave cavity. To determine the amount of H produced, the fast titration reaction $H + NO_2 \rightarrow NO + OH$, with a rate of $1.3.10^{-10}$ cm^3/s.molecule, is used [5]. Calibration of the N-density, although less straightforward, can be done in a similar way. The absolute number densities of the atomic species are not only important to gain information on the fundamental plasma processes and conditions, like the degree of dissociation, but also to obtain a quantitative idee of the sensitivity of the TALIF technique for the various atomic species.

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