CONFERENCE PRE-PRINT

ACTIVE SPECTROSCOPY FOR ATOMIC H AND D MEASUREMENTS IN FUSION

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Abstract

Atomic hydrogen (H) and deuterium (D) are key species in fusion plasmas, influencing ionization, radiation, recombination, and plasma–wall interactions that govern divertor and scrape-off layer (SOL) behaviour. However, quantitative measurements of H and D in fusion devices typically rely on indirect methods based on optical emission spectroscopy (OES) and complex models to interpret OES data. An interesting alternative is two-photon absorption laser-induced fluorescence (TALIF), a technique that is capable of direct and localized measurements of the absorption spectrum of H and D. We have implemented a picosecond TALIF diagnostic in the RAID linear plasma device at the Swiss Plasma Center. The system enables spatially resolved (1D) measurements of H density in hydrogen plasmas, surpassing previous indirect spectroscopic approaches. The results demonstrate the value of TALIF for fusion-relevant plasma studies. Furthermore, a femtosecond (fs-)TALIF system has been developed to investigate single laser pulse determination of H and D densities in RAID. Individual fs-pulses can excite H and D populations at the temperatures expected in divertors and SOLs, allowing for measurements better suited to tokamak operations. Fs-TALIF may open new possibilities to diagnose neutral species in tokamaks and other high-power devices.

1. INTRODUCTION

Atomic hydrogen (H) and deuterium (D) play a fundamental role in fusion plasmas, particularly in ionization, line radiation, and recombination processes, which significantly influence the particle, momentum, and energy balance in divertors and scrape-off layers (SOLs) of fusion devices. Additionally, H and D interact with wall surfaces, affecting adsorption, desorption, and recycling, which in turn shape divertor and SOL dynamics. Despite their importance, quantitative measurements of H and D in tokamak SOLs and divertors remain scarce, partly due to the operational constraints of tokamaks. To address this, we have used the flexibility and diagnostic accessibility of the Resonant Antenna Ion Device (RAID) at the Swiss Plasma Center (SPC) to develop and operate a two-photon absorption laser-induced fluorescence (TALIF) system using a picosecond (ps-) laser. This system enables high-resolution 1D-resolved measurements of H density in hydrogen plasmas that could previously only be performed using indirect spectroscopic methods and modelling. Our experiments demonstrate the usefulness of the technique in fusion-relevant studies and open the door to investigations of its applicability in high power devices using a fs-laser system, presently under development.

2. THE RESONANT ANTENNA ION DEVICE (RAID)

RAID [1] is a linear (1.5 m total length and 0.4 m diameter) basic plasma physics device in operation at SPC. In RAID, steady-state helicon plasmas in various gases, including H_2 and D_2, are produced by two radio frequency (RF) birdcage resonant antennas [2] delivering a maximum power of 10 kW per antenna at 13.56 MHz. The cylindrical vacuum vessel is water-cooled and allows for long-time (up to days) continuous operation with stable and reproducible plasma conditions. Seven magnetic field coils, surrounding the vacuum chamber, generate an axial field of up to 660 G on axis. Typical electron density in H_2 and D_2 plasmas can be as high as 7×10^{18} m^{-3} on the axis of the column with good homogeneity along the axial direction. Typical on-axis electron temperatures are ~1-7 eV. Parameters such as the neutral gas pressure, the RF injected power and the background magnetic field can be varied to control plasma profiles and access a variety of plasma conditions of relevance for tokamak divertor studies.

A schematic of RAID with a few selected ancillaries is shown in Fig. 1. Each magnetic coil can be independently powered by individual power supplies allowing the production of expanding magnetic field configurations, cusp field, etc. Each coil is made of 9 (radial) × 4 (axial) turns with a central channel for water cooling. Each turn is electrically isolated with resin. The coils are actively cooled by forced water convection in the copper pipes for continuous operation. The coil internal and external diameters are 52 cm and 82.6 cm, respectively. The polarity

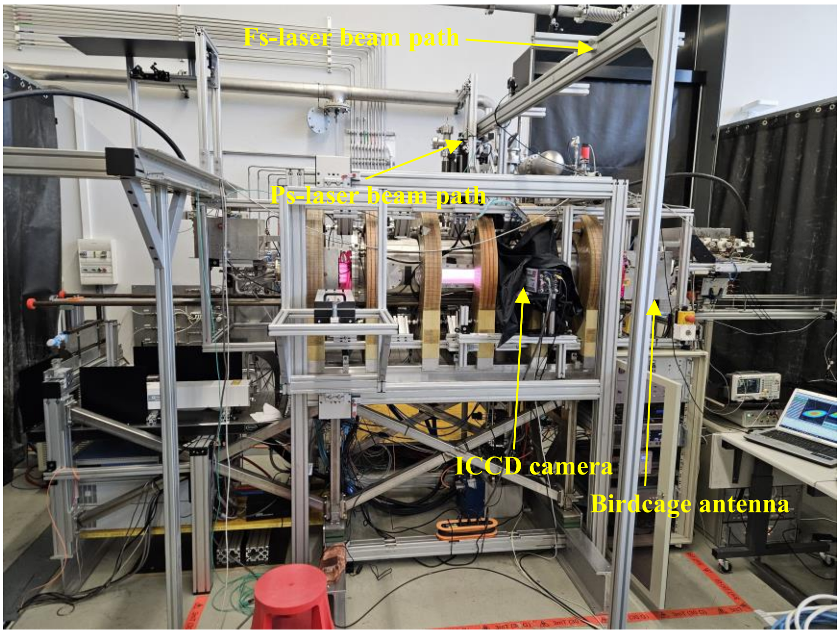


Figure 1: Picture of the RAID device with ancillaries. The laser beam paths (for both the ps-laser and the fs-laser) as well as the ICCD camera for detection of the fluorescence light are shown.

of the coils close to the plasma source can be reversed with respect to the adjacent coil.

The birdcage resonant antennas are mounted axially on both ends of the vacuum chamber. Each one is made of 9 conducting parallel legs distributed around a dielectric tube in a cylindrical configuration. The antenna diameter is 13 cm and the 15 cm-long conducting legs are made of copper tubing for water-cooling of the system to allow for continuous operation. Each leg is connected at both ends to its closest neighbours by an assembly of 10 high-Q ceramic capacitors in parallel, rendering the system resonant at 13.56 MHz. This novel antenna design represents an alternative to conventional partial-helix helicon antennas and can maintain a stable and intense steady-state helicon plasma column [2, 3] with advantages of wider parameter space and improved plasma stability, especially for light-ion (H/D) plasmas. The helicon plasma is sustained with a minimum power of ~ 1 kW in H_2 and D_2. The plasma length is approximately 2.10 m between the two antennas. Plasmas exhibit a peaked radial profile of T_e and n_e, typically with a few centimeter radial half-width, which depends mostly on the magnetic field and RF injected power.

RAID is equipped with basic plasma diagnostics as well as sophisticated state-of-the-art systems, to characterize the different plasma components (electron, ions, molecules...) as well as magnetic fields associated with helicon waves. The relatively low temperatures and densities of RAID plasmas, in some regimes, allow the use of Langmuir probes (LPs) to measure the electron density and temperature profiles. LPs are mounted on a two-axis system, which can scan axially and radially inside the plasma column to measure the full plasma profile [4, 5]. Single tip LPs, RF-compensated LPs, as well as double tip LPs are used depending on the plasma conditions. However, LPs suffer strong heating for an electron density larger than a few 10^{18} m^{-3}, which is achieved in argon at relatively low RF power (> 300 W), and in hydrogen or deuterium at RF power > 3000 W. This restricts the use of LPs to the edge of the plasma column depending on the gas and plasma regime. To overcome this limitation, microwave interferometry and incoherent Thomson scattering (TS) are used in parallel with LPs to probe all RAID plasma regimes as well as to extract calibrated electron density profiles.

TS is used to measure the electron velocity distribution function, thereby providing the electron temperature as well as the calibrated electron density. The TS setup operates in the incoherent scattering regime, where the photon energy is altered solely due to the Doppler shift by the free electron velocity in the plasma along the probe vector. A first system used polychromators and a 1064 nm laser [6]. The improved TS system uses a spectrometer and a 532 nm laser focused to 1 mm beam diameter. The scattered radiation is collected with a spatial resolution of 4 mm along the laser beam at a 90° scattering angle and is guided to a high-throughput spectrometer (FL = 200 mm, 2400 l/mm) by an optical fiber. A photo multiplier tube with a rise time of 0.57 ns detects the scattered light. The spectrum is scanned by varying the spectrometer grating angle. The setup has a spectral resolution of 0.8 nm and can therefore measure electron temperatures > 0.4 eV. A similar TS system has been successfully used on the Helicon Plasma Source (HPS) at AWAKE [7].

3. TWO-PHOTON LASER-INDUCED FLUORESCENCE (TALIF) TECHNIQUE

Laser-induced fluorescence (LIF) is a spectroscopy technique commonly employed to determine the velocity distribution function (VDF) of atomic and ionic species, particularly in low-temperature plasmas. As an active diagnostic, LIF enables direct and spatially resolved measurements of the absorption spectrum of the target atoms.

Once effects such as Zeeman or Stark broadening are considered, the measured spectrum provides access to the VDF of the absorbing species. The VDF then allows to determine the species' temperature and flow velocity. Under certain conditions, determination of the absorption spectrum also allows to establish the density.

In fusion research, single-photon LIF schemes have been applied to determine hydrogen densities, for example in the TEXTOR device [8]. To probe hydrogen in its ground state, a vacuum ultraviolet (VUV) beam tuned to the Lymann α transition at 121.5 nm ($n = 1 \rightarrow n = 2$) was used. Detection of the corresponding Lymann α fluorescence then allowed reconstruction of radial profiles for both density and temperature of ground-state hydrogen. Despite their utility, such LIF schemes face major limitations. The generation and manipulation of VUV radiation are technically demanding, as they require specialized optics and are strongly absorbed by most materials as well as in the plasma edge region, where neutral particle densities can be high. Additionally, the suppression of stray laser light necessitates complex optical arrangements. Altogether, these requirements impose stringent challenges on the practical implementation of LIF diagnostics in fusion devices.

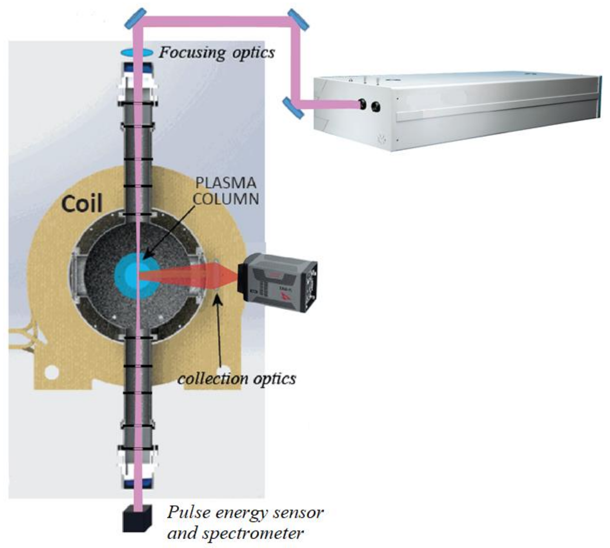


Figure 2: The ps-TALIF setup with an end-on view of RAID. The main chamber, coils and hydrogen plasma column are schematically shown. The laser beam is also shown to indicate the axial position of the TALIF collection optics mounted on a lateral window. The main elements of the TALIF system are shown including the field of view of the ICCD camera across the plasma column and the laser beam, which terminates on a pyroelectric detector used for the measurement of the pulse energy.

An alternative LIF technique for hydrogen, first proposed in Ref. [9], uses two-photon absorption to excite ground-state atoms into the n=3 levels. A fraction of these excited atoms then decay to n=2, releasing fluorescence. In this method, two photons at 205.1 nm—each carrying half of the transition energy—are absorbed simultaneously. This bypasses the restrictions of single-photon vacuum ultraviolet (VUV) excitation but yields a much weaker signal, since the two-photon absorption cross-section is orders of magnitude smaller. As a result, high-power, short-pulse lasers (typically on the nanosecond scale or shorter) are needed to generate detectable fluorescence. Because the energy spacing between the n=3 states is small compared to the laser bandwidth, multiple states can be excited. However, most fluorescence is expected to originate from the 3d state, with only a minor contribution from 3s. Direct two-photon excitation to 3p is forbidden by angular momentum conservation, and the cross-section for 3s is significantly smaller than that of 3d. Furthermore, the spontaneous emission rate from 3s → 2p is much weaker than from 3d → 2p. Taken together, these considerations indicate that fluorescence arises predominantly from the 3d state, which has a natural lifetime of about 15.5 ns.

Over the past few decades, such two-photon absorption laser-induced fluorescence (TALIF) diagnostics have been applied to the study of hydrogen plasmas, especially in the context of atmospheric plasmas [10]. An advantage of TALIF of hydrogen is that an absolute calibration procedure is possible using krypton gas at a known pressure and temperature. The method relies on the closeness in wavelength between the two-photon excitation of ground-state atomic hydrogen (205.1 nm) and ground-state krypton (204.1 nm). The calibration is performed by recording the TALIF signal from krypton and computing the ratio with the hydrogen TALIF signal acquired under identical optical and detection configurations. Corrections are applied for differences in excitation wavelength, detection efficiency, and quenching rates. This approach provides a reliable means of converting TALIF intensities into absolute hydrogen atom densities in plasmas. An overall ~50% uncertainty in the cross-section ratio between hydrogen and krypton is usually the limiting factor in the accuracy of TALIF measurements of absolute H density.

On RAID, a ps-TALIF system [11] has been in operation in the past years and a fs-TALIF system is presently under commissioning with the final goal of assessing the feasibility of the TALIF technique in fusion devices. Both TALIF systems with their experimental results are described in the following.

4. ATOMIC H OR D MEASUREMENTS ON RAID USING PS-TALIF

The ps-TALIF system on RAID [11] is schematically shown in Fig. 2. The UV pulses employed in the ps-TALIF system are generated by an EKSPLA ps-laser system, consisting of a PL2231 50-TRAIN pump laser operating at 1064 nm, coupled to a third-harmonic generator and a PG411 SH-DUV optical parametric generator. This configuration provides tunable radiation in the 193–2300 nm range, with a 50 Hz repetition rate and a pulse energy up to 100 µJ in the 204 - 205 nm range used for the present studies. The ~28 ps pulse duration is much shorter than the ∼ns timescale of the collisional and radiative processes in the plasma. The bandwidth of the pulses is expected to be $\leqslant$ 5 cm−1, which corresponds to $\leqslant$ 0.02 nm for the UV pulses used in our experiments. The wavelength can be tuned in increments of 0.01 nm, which is comparable to or larger than the Doppler width of hydrogen atoms—on the order of 10^{-3} nm for room-temperature atoms and 10^{-2} nm for plasmas near ~1 eV. Although no absolute calibration of the laser wavelength was carried out (with an accuracy likely limited to a few × 0.01 nm), the tuning steps are highly precise, enabling spectral scans that resolve the laser absorption profile.

The UV beam is directed into the RAID chamber along a ~10 m optical path using a sequence of seven mirrors, entering from the top of the device (see Fig. 2). To maximize transmission efficiency, the mirrors (Laseroptik GmbH) are custom-made with high-reflectivity coatings (R > 0.99) at 205 nm. The beam is then focused using a lens system to an estimated 280 µm diameter at the focal point near the vessel axis, corresponding to a Rayleigh length of XR ≈ 30 cm. These parameters were optimized to enhance the TALIF signal while avoiding excessive focusing, which could otherwise saturate the fluorescence emission. After traversing the RAID vessel, the laser beam is terminated on a pyroelectric detector (Gentec QE8SP-B-MT-D0), allowing the measurement of the energy of individual pulses. The fluorescence generated after two-photon absorption of the laser beam is collected by a set of lenses into a Princeton PI-MAX 4 ICCD camera mounted perpendicularly to the laser beam.

The measured, spatially resolved, decay times for H are shown in Fig. 3-Left. TALIF measurements of H are performed in a steady-state plasma generated in RAID with a 4 kW of RF power, a magnetic field of 325 G on-axis, and an H_2 fill pressure of 1.59 Pa. In these conditions, the electron density and temperature are 10^{18} m^{-3} and 1eV, respectively, at the center of the plasma column (close to the axis of RAID) and radially decreasing at the edge 2×10^{17} m^{-3} and 0.75 eV, as determined in separate measurements using TS [10]. Calibration with Kr was carried out under no plasma conditions, with the RAID vessel at room temperature (≈ 300 K) and at a fill pressure of 8.95 Pa for a uniform density 2.16×10^{21} m^{-3}. This avoids possible undesired effects such as quenching from collisions with neutrals or amplified spontaneous emission.

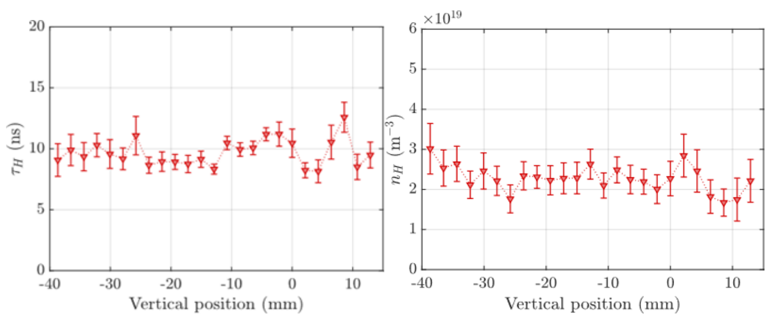


Figure 3: Left – Profile of fluorescence decay times of H across the dense hydrogen plasma column of RAID (axis of device at 0 mm). Right - H absolute density profile.

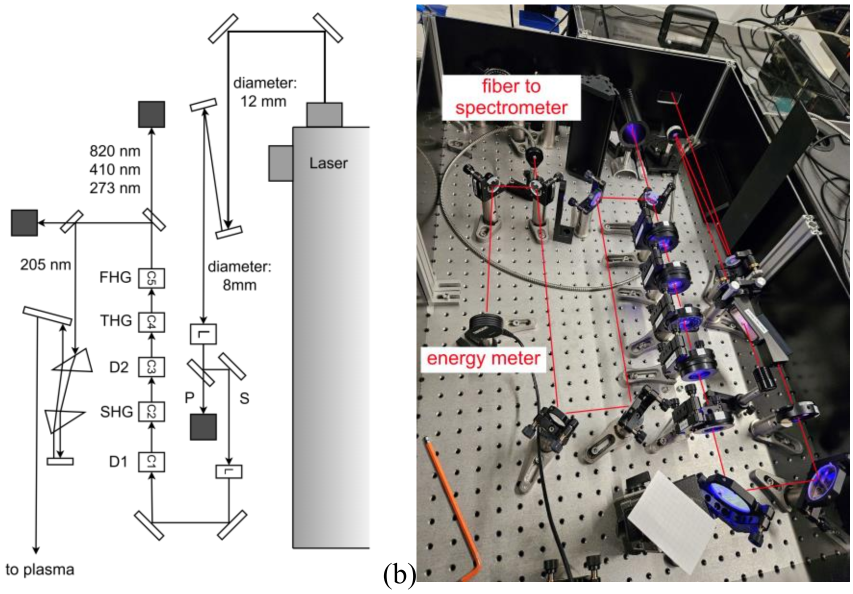
The absolute H density profile determined in these experiments is consistent with a flat profile across the cylindrical plasma column of RAID, up to a radius of 4 cm, a region over which the plasma density and temperature show a significant variation. The average H density across the measured region is (2.3 ± 0.1) $\times10^{19}$ m^{-3}. This corresponds to an average dissociation degree of ~2.9%. The spatial, 1D-resolved, profile is consistent with a uniform decay time across the measured region, for both H and Kr (not shown, for details, see Ref. [10]). In the case of H, the profile has a mean of 9.7 ns +/- 0.2 ns, which is significantly lower than the purely optical decay time of the 3d state, estimated to be ~15.5 ns. Electron impact excitation rates are not sufficiently large to quench the excited state at a sufficiently high rate, suggesting that other processes play a significant role. One particularly interesting possibility is the occurrence of complete substate mixing of the n=3 states of hydrogen since it yields decay times consistent with the observations at all locations (see Ref. [10]).

Accounting for complete mixing requires a change in the determination of the branching ratio as well, possibly leading to a 26% increase in the H density measurements. Further theoretical and experimental studies are nevertheless required to better understand these observations.

5. DEVELOPMENT OF A FS-TALIF SYSTEM FOR RAID

Traditional ns- or ps-lasers have a narrower spectral width than the expected absorption linewidth of H and D in the SOL or divertor. The wide absorption width arises from broadening mechanisms such as Doppler shifts of atoms with kinetic energies up to ~10 eV, Zeeman splitting at ~1 T magnetic fields, and possibly Stark broadening. As a result, ns- and ps-laser-based systems require scanning the laser wavelength across the entire absorption line to obtain the spectrally integrated fluorescence signal needed for density calculations, something that is not possible over the time scale of typical tokamak discharges (which typically last a few seconds).

Femtosecond (fs-)pulses provide an attractive alternative to overcome this critical limitation of earlier TALIF methods for tokamak applications. Fs-laser pulses have large spectral widths of the order of 0.1–1.0 nm, capable of exciting in each pulse the local H and/or D ground-state populations independent of the expected laser absorption broadening mechanisms. Fs laser pulses may therefore enable single-laser pulse measurements of density, eliminating the constraint of performing scans and allowing to capture the rapidly changing conditions of fusion plasmas. We are presently developing, as part of a EUROfusion ENR project [12] led by Dr. M. Baquero-Ruiz, a fs-TALIF system to investigate single-laser pulse determination of H/D densities in RAID to explore the



(a) (b)

(b)

Figure 4: The schematic (a) shows the layout of the fourth-harmonic generator of the fs-TALIF system, and the picture (b) shows the implementation in the laboratory.

potential as well as possible limitations of fs-TALIF to diagnose fusion-relevant plasmas. The design of the fs-laser system is reviewed below.

Leveraging the results with the ps-laser system, we target the wavelength of 205.1 nm for TALIF in hydrogen. For this, we have designed a fourth harmonic generator (FHG) based on sum-frequency generation to obtain the required wavelength starting from ~820 nm. We follow the design of Susnjar et al. [13], who created a novel FHG based on five non-linear crystals in a common laser path. This design was selected because of its relative simplicity and expected stability. In Fig. 4, the schematic (a) shows the layout of the FHG, and the picture (b) shows the implementation in the laboratory. More technical details can be found in Refs. [14, 15]. An Astrella fs-laser, manufactured by Coherent, generates a laser beam at 820 nm with a pulse duration of 70 fs and a pulse energy of 7 mJ. The laser system has a repetition rate of 1000 pulses per second. First, the diameter of the beam is reduced by using a reflective telescope to achieve high wavelength conversion efficiency. Second, a device consisting of a waveplate and two Brewster windows is used to set the laser energy for the experiments. This device only outputs the S-polarised part of the incoming beam. By rotating the waveplate, the energy of this outgoing beam can be selected. Third, the beam propagates through another waveplate and five non-linear crystals. These crystals are used for the harmonic generation. The second harmonic-generating (SHG) crystal converts 10-20% of the incoming beam into the second harmonic at 410 nm. The third harmonic-generating crystal (THG) combines a part of the incoming 820-nm and 410-nm light and converts it into the third harmonic at 273 nm. Two delay plates (D1 and D2) are needed for this step. The FHG crystal combines a part of the incoming 820 nm and 273 nm light, to create the fourth harmonic at 205 nm. These conversion steps are dependent on the beam polarization, and thus an initial waveplate is required. Fourth, as all crystals lie in the same beam path, a series of monochromatic mirrors is used to isolate the 205 nm beam.

The last part of the FHG is the pulse compressor, which is based on two prisms. The goal of the pulse compressor is to compensate for the dispersion in air that the fs pulse may experience when propagating from the laser to the RAID device. After the prisms, the laser is sent through a sealed beam path towards RAID for injection and propagation through the plasma. The pulse compressor is not yet installed in the present setup, as it was found that the power loss in the prisms is considerable (20% per prism-traversal, i.e. 59% in total), but experiments are planned with it to test the effect of dispersion of the fs pulses in the fluorescence signal.

The FHG can thus generate high-energy wide-bandwidth 205 nm pulses. The fs UV laser pulses are routed to RAID using the sealed beam path which, if needed, can be put under vacuum. We minimize refractive elements in it to (1) reduce pulse dispersion and (2) to avoid damage due to absorption of the high intensity pulses. At our pulse energies, the beam can be focused loosely to use the full energy while avoiding saturation.

6. CHARACTERIZATION OF FS LASER PULSES

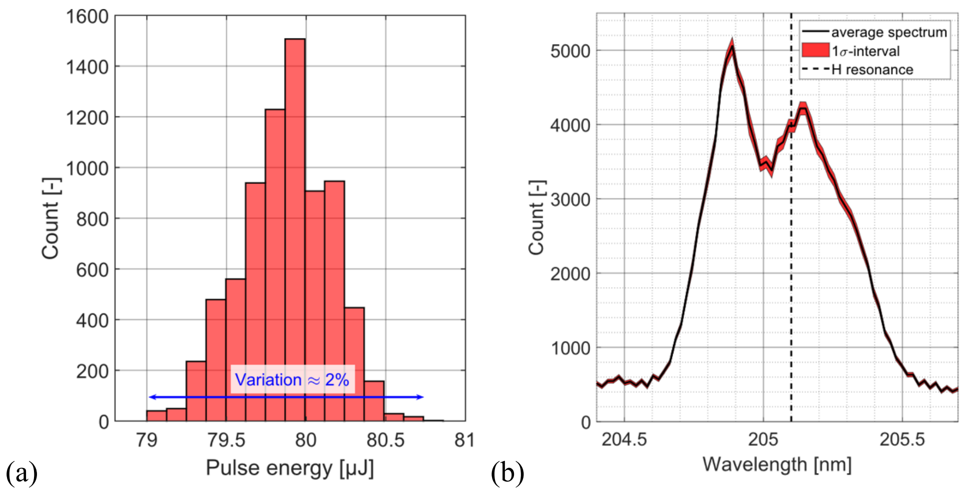


Figure 5: First characterization of the fs-laser pulse. (a) Energy stability: energy distribution of laser pulses; (b) Spectral stability.

The pulses generated by the FHG were analyzed in terms of their energy and spectral stability. The energy of the 205 nm beam was measured using a Coherent J-10MT-10KHZ energy sensor, positioned at the end of the laser path (Fig. 4). To protect the sensor from damage, the 205 nm beam was isolated from the other harmonics by means of DUV mirrors. The small fraction of 205 nm light transmitted by these mirrors is exploited to record the pulse spectrum. For this, a UV fiber is placed behind the fifth mirror, ensuring that both the fiber and the connected Avantes UV spectrometer were shielded from the residual power of the other harmonics. Pulse stability is assessed by recording the energy and spectrum of 8000 consecutive pulses. The energy distribution (Fig. 5-a) follows a Gaussian profile with an average of 79.9 μJ and a 3σ variation of ±1.0 % (half-width-half-maximum), confirming the excellent stability of the 205 nm pulse energy. The spectral results (Fig. 5-b) show an average spectrum centered at 205 nm with a FWHM of 0.6 nm and a maximum deviation of ±6.7 % from the mean, indicating high spectral stability as well. The combination of high pulse energy and broad spectral width is a key outcome, as both are essential for single-laser-pulse TALIF experiments. Even though the observed spectrum deviates from a Gaussian shape, recent work to optimize the laser has led to improved spectral qualities of the pulses at the expense of reduced pulse energies. Investigations are ongoing to find the best compromise between the two quantities.

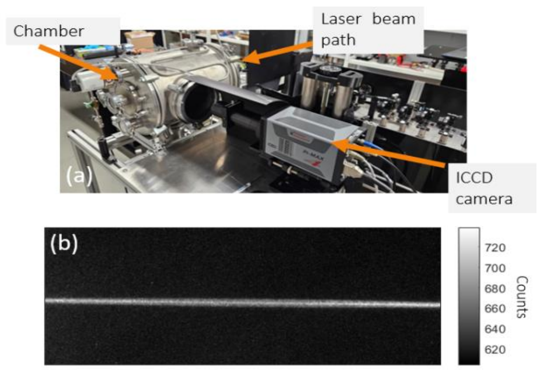


Figure 6: (a) Setup for measurements of fs-TALIF in Kr gas. (b) Fluorescence upon two-photon absorption in Kr, as captured by the ICCD camera. The signal is observed along a ~5 cm long path.

6. FS-TALIF IN Kr GAS

First experiments with the fs-system have recently been carried out in Kr gas, using a small chamber at room temperature at a fill pressure of 30 Pa. The experimental setup is shown in Fig. 6. The fs-laser beam is collimated and injected along the axis of the cylindrical chamber (see Fig. 6-top), and fluorescence stemming from TALIF is detected along the laser path on a circular window using an ICCD camera capable of fast gating.

With the laser wavelength set at 205.1 nm, fluorescence is observed along the entire path of the laser within the field of view of the detection system (~5 cm), as shown in Fig. 6-bottom. Since the Kr two-photon transition is resonant at 204.1 nm, this observation shows that the wide spectral width and high energy of the pulses is sufficient to excite Kr, enabling possible absolute calibrations without the need of scanning wavelength. Further investigations into the applicability of this technique are ongoing.

7. CONCLUSIONS AND OUTLOOK

We have applied TALIF with ps-pulses to determine one-dimensional profiles of H ground-state densities in RAID plasmas relevant to the study of tokamak scrape-off layers (SOLs) and divertors. A major effort has been devoted to quantifying uncertainties to improve the reliability of both the diagnostic system and the measurements. The results obtained are consistent with a uniform ground-state H density across the measured region, which corresponds to the plasma column of RAID, a region over which significant variation in plasma density, as well as temperature, is expected.

From these first measurements, we determine an average dissociation rate of hydrogen of 2.9 %, which increases to 3.7 % (a 26 % difference) when the effect of full H sub-state mixing is included. Furthermore, the measurements of fluorescence decay are compatible with full mixing across the plasma column. Ongoing studies aim to better characterize this process and to determine how it should be correctly incorporated into models to interpret TALIF signals.

Experimental improvements of the ps-system are underway. We are currently enhancing the optical system, detection scheme, and beam focusing to increase the signal over noise ratio. Future work will include the investigation of denser and hotter plasmas.

In parallel, we are implementing TALIF of H using fs-pulses within a EUROfusion Enabling Research project, in collaboration with the Lausanne Center for Ultrafast Science (LACUS) at EPFL. We have developed an fs-system capable of delivering stable, high energy and broad spectral-width pulses required for single-laser-pulse TALIF generation and detection. TALIF in krypton gas has been demonstrated using pulses tuned to the hydrogen resonance. We expect to inject the fs-laser beam into RAID shortly and to begin plasma experiments. Alongside these developments, significant theoretical and experimental work has been carried out on fs two-photon absorption as well as on hydrogen substate mixing, strengthening the basis for future investigations.

We believe that the fs approach offers promising opportunities for future applications in tokamak environments.

ACKNOWLEDGEMENTS

This work has been carried out within the framework of the EUROfusion Consortium, via the Euratom Research and Training Programme (Grant Agreement No 101052200 —EUROfusion) and funded by the Swiss State Secretariat for Education, Research and Innovation (SERI). Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union, the European Commission, or SERI. Neither the European Union nor the European Commission nor SERI can be held responsible for them.

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