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Development of pulsed UV lasers and their application in laser spectroscopy

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Development of pulsed UV lasers and their application in laser spectroscopy

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Abstract. The application of two-photon laser spectroscopy to plasma diagnostics requires tuneable UV-laser spectrometers providing: some mJ pulse energy at ns time scale with spectral quality close to Fourier Transform Limit, good pulse to pulse reproducibility and tuning linearity. We report about two different systems, a first laser specially optimized for the radiation at 243 nm, which is required for the 1S-2S two photon transition of atomic hydrogen, and a second one generating 205 nm suited for the transition 1S - 3S/3D.

1. Introduction

The main goal of experimental plasma physics is to obtain a complete characterization of the plasma state; this means the determination of plasma parameters such as particle density, temperatures, electric field strength etc. One significant parameter in the physics of discharges is the local electric field strength (E-field), because it determines the charged particles fluxes and their energy distributions. The knowledge of this magnitude is relevant for understanding the plasma discharge and improving modeling. For this reason, many techniques have been developed for E-field determination, among these techniques non intrusive methods, like laser spectroscopy, are preferred [1, 2].

We have determined the E-field in the cathode fall region of a hollow cathode discharge, by two laser spectroscopy techniques: Doppler-free two-photon optogalvanic spectroscopy and Doppler-free two-photon polarization spectroscopy via the Stark splitting of the 2S level of atomic deuterium and hydrogen for a wide range of discharge conditions (currents from 50 to 200 mA and pressures from 400 to 1350 Pa) [3-5]. The Stark effect, i.e., changes in the atomic spectra depending on the E-field, results in shifting and mixing of spectral components and variations in their intensities. Comparisons of the measured spectra with theoretical calculation provide the E-field. Measurements down to 400V/cm have been done. We plan to improve the two-photon optogalvanic spectroscopy as a tool to measure strong E-fields, and its further application for studying the cathode fall characteristics in hollow cathode discharges. The detection sensitivity can be improved by at least one order of magnitude, using Doppler-free two-photon optogalvanic spectroscopy applied to the 1S - 3S/3D transition of hydrogen isotopes instead of the 1S-2S transition.

However, all of these measurements require tunable UV-laser spectrometers, providing sufficient pulse energy with a very narrow spectral bandwidth, good pulse to pulse reproducibility and scan linearity. Because commercial laser systems can not provide such a radiation, we started more than one decade ago to develop advanced pulsed single longitudinal mode, SLM UV-laser spectrometers, especially suited for laser-aided plasma diagnostic [6-8].

2. UV-laser spectrometers

Tuneable laser radiation at 243 nm suited for two-photon spectroscopic measurements at the 1S-2S transition of hydrogen, is provide by: a 10 Hz injection-seeded Q-switched Nd:YAG laser (Continuum, Powerlite 8000), a substantially modified system (Continuum, Mirage 500, which is not longer commercially available) consisting of an Optical Parametric Oscillator (OPO), followed by an Optical Parametric Amplifier (OPA), and finally Sum Frequency Generation (SFG) in a BBO crystal. In detail, in the first step, the second harmonic of the Nd:YAG laser at 532 nm is split into tunable signal and idler wave by a Type II KTP-OPO operated at SLM in the near-infrared radiation (signal wave at 772 nm). In a second parametric process the signal wave is amplified in an OPA, based on two Type I BBO crystals, pumped with the third harmonic of Nd:YAG at 355 nm. According to our concept this system is extended to produce UV-radiation by mixing the OPA output with the leftover of the third harmonic pump beam in a SFG process in an additional Type I BBO crystal. The whole chain of non-linear processes involved had to be optimized for high conversion efficiency, stability and reliable operation of the system. This concept provides up to 10 mJ pulse energy in 2.5 ns and 300 MHz bandwidth at 243 nm with excellent performance. The beam divergence is less than 100 μrad and the pointing stability during hours is better than 50 μrad. The pulse energy conversion efficiency from the infrared output of the Nd:YAG laser into tunable UV-radiation is about 2%. Since the OPO reduces the pulse duration by a factor of three, the pulse peak power conversion efficiency is as higher as 5%.

In order to apply such a system to two-photon laser spectroscopy for plasma diagnostics is important to verify the quality of the laser emission while recording the spectra. To be sure that the tuning of the laser in frequency is regular, the OPO signal (at 772 nm) is controlled by a monitor etalon with free spectral range (FSR) about 7 GHz and a photodiode. A typical set of the fringes obtained during a complete scan can be seen in figure 1. From this, we can derive that the regularity of the frequency detuning is typically about 1 % of the FSR of the etalon during the scans we are usually doing for spectroscopy measurements, and has been 4 % in the worse case during all measurements in the same campaign.

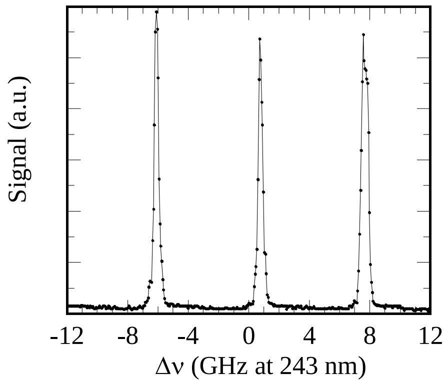

Figure 1: Fringes of a monitor etalon as a control of the regularity of the laser frequency detuning

An optogalvanic cell, containing hydrogen at a pressure of about 500 Pa, provides the bandwidth control of the laser, and serves as frequency reference for the 1S-2S transition. Partial thermal dissociation of the molecules is obtained by a tantalum wire resistively heated to a temperature of about 1500 K. Less than 50 μJ pulse energy are sufficient to provide Doppler-free two-photon excitation by counter propagating beams. Subsequent ionization by absorption of a third photon produces charged particles, which are finally measured by a pickup electrode as a current pulse. An example of the optogalvanic signal is given in figure 2. The signal is fitted to the sum of two Lorentzian profiles, and exhibits clearly the hyperfine structure splitting of 621 MHz of the 1S-2S hydrogen resonance. This shows that the upper limit of the laser bandwidth is about 300 MHz.

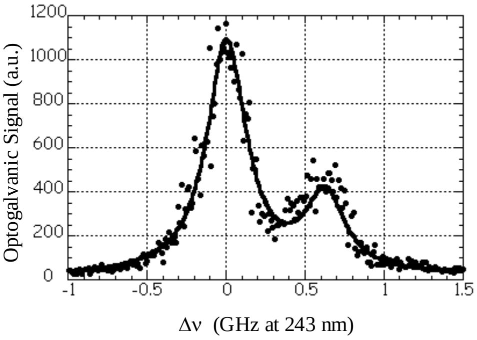

Figure 2: Hyperfine splitting of the 1S-2S hydrogen resonance as a control of the narrow laser bandwidth

As result of our experience in sensitive spectroscopic techniques and lasers suitable for plasma diagnostics, our group had developed a pulsed laser system generating high power SLM tunable radiation at wavelengths down to 200 nm. The system is specially optimized for the radiation at 205 nm, which is required for the 1S - 3S/3D two photon transition of atomic hydrogen. The pulsed laser system is based on a similar modular concept as the laser at 243 nm. The 205 nm radiation is generated by a Nd:YAG laser (Continuum, Powerlite 9000), a seeded KTP-OPO, two Ti:sapphire amplifiers, and by stepwise SFG via Second Harmonic Generation (SHG), Third Harmonic Generation (THG) and Four Harmonic Generation (FHG) using BBO-crystals.

The short resonator consists of a KTP crystal placed in a plane mirror optical cavity, pumped by the second harmonic of the Nd:YAG laser . SLM pulsed operation is achieved by seeding with a tunable external cavity cw diode laser. The special feature of the OPO is its linearity frequency tuning performed by changing the cavity length in a controlled way: one resonator mirror is mounted on a translator stage with nanometer precision, and the diode laser is frequency locked to one cavity mode. The pulse energy of the OPO output at 820 nm is about 1 mJ, and tunable over a range of about 50 GHz.

The OPO pulse is amplified in two Ti:sapphire crystals pumped also by the second harmonic of the Nd:YAG laser. The first Ti:sapphire crystal provides six pass amplification while the second crystal serves as two pass amplifier, generating pulse energies of about 60 mJ. Finally, in order to obtain the best conversion efficiency, the amplified 820 nm radiation is converted into the UV by stepwise SHG, THG and FHG using three BBO crystals. The radiation at 205 nm has up to 5 mJ pulse energy in about 4 ns and a spectral bandwidth around 300 MHz. In addition to the high pulse energy and the narrow bandwidth, the system provides good pulse-to pulse reproducibility and excellent scan linearity.

3. Conclusion

We have demonstrated the utility of a high power SLM UV-laser spectrometer (some mJ pulse energy, 2.5 ns pulse duration, 300 MHz bandwidth) for plasma diagnostic based on Doppler free two-photon spectroscopy (243 nm) via the 1S-2S transition of hydrogen isotopes. The generation of tuneable UV radiation is based on a modular concept: conversion of the second and third harmonic of a commercial injection seeded Nd:YAG (Q-switched) into tuneable SLM radiation at 772 nm using a

KTP-OPO, amplification by a BBO-OPA and final UV generation by SFG of the 772 nm with the third harmonic of the Nd:YAG. The 300 MHz bandwidth allows e.g. to resolve the hyperfine structure of hydrogen and the tuneability is sufficient for measuring easily the electric field strength (larger than 1 kV/cm) in the cathode fall of hollow cathodes discharges operated with hydrogen isotopes. The field strength sensitivity can be increased at least by one order of magnitude using SLM radiation of 205 nm and the 1S-3S/D transition of hydrogen. For this we are setting up another modular SLM UV laser spectrometer realising the following concept: conversion of the second harmonic of a commercial injection seeded Nd:YAG (Q-switched) into tuneable SLM radiation at 820 nm using a novel seeded KTP-OPO, amplification in TiSa (also pumped by the second harmonic), and stepwise conversion SFG of the 820 nm radiation via SHG, THG and FHG using BBO-crystals. The novel seeded OPO concept provides a tuning range of 200 GHz at 205 nm with short (large) range precision better than 80 MHz (400 MHz) as required for the precise electric field strength measurements.

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