A portable diagnostic system for the quantification of VUV fluxes emitted from low-temperature plasmas

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Abstract. Vacuum-Ultraviolet (VUV) spectroscopy is linked to a huge effort, in particular if absolute numbers are required. To overcome this, a flexible device is developed based on a photodiode and optical filters for wavelength selection. Characterization of the diagnostic is performed against a VUV spectrometer that is radiometrically calibrated down to 46 nm. In the first instance, the latter is used for an analysis of VUV spectra of inductively coupled low-temperature plasmas for a variety of discharge gases and mixtures. The measured photon fluxes can easily have comparable magnitude as the occurring ion flux, and it is demonstrated that the photonic energy distribution can change drastically with varying external parameters (pressure, RF power). This highlights the need for energy resolution for VUV flux detection and a set of ten optical filters was selected for the VUV diode system according to the respective prominent emission ranges of typical discharge gases for plasma processing. Recommended filter combinations for each of the discharge gases and mixtures are given. The developed diagnostic is calibrated in-house against the VUV spectrometer, which makes energy-resolved absolute VUV flux measurements up to photon energies of 27 eV possible. The calibration is performed individually for all the investigated discharge gases and the corresponding filter combinations, and an accuracy of better than 25 % compared to the VUV spectrometer is attested for the investigated parameter space (0.3–10 Pa, 200–1100 W RF power). Its applicability is demonstrated by measuring VUV fluxes at two further low-temperature plasma setups.

Keywords: VUV emission, photon flux, photodiode, low-temperature plasma, synthetic air, gas mixtures

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1. Introduction

Vacuum-ultraviolet radiation (VUV) is characterized by wavelengths of less than 200 nm, which implies energies of at least 6.2 eV per photon. In low-temperature plasmas, such radiation is predominantly present due to resonance transitions between the ground and the first excited states of neutrals and ions. Evaluating this emission thus grants access to basic plasma physics phenomena. Furthermore, surfaces immersed in such plasmas are inherently exposed to the hard UV radiation and due to the high energies involved, their contribution to plasma-surface interaction is of high relevance [1–6]. Moreover, several studies have already shown that photon fluxes can be of the same magnitude as ion fluxes [5, 7, 8], and that the involved photon energies can determine the impact on the surface due to possible energetic thresholds [1, 6] and the correlated penetration depth [1, 2]. Hence, measuring VUV fluxes wavelength-resolved is indispensable for characterization and tuning of the plasma in view of a certain application.

The straightforward solution is to use VUV spectrometers. From 200 nm downwards, strong absorption bands of oxygen exist, which requires that VUV detection systems are connected to the vacuum system of the plasma under observation. For the typically heavy instruments, this is not always possible. Alternative solutions, mostly developed in the context of plasma processing, are commonly based on a VUV-sensitive photodiode [9, 10] or a phosphor (sodium-salicylate) to convert the VUV radiation to visible radiation [9, 11]. In both cases, wavelength resolution can be obtained by applying optical filters [9–11]. More sophisticated approaches include chemical dosimeter-type detectors [12] or self-built sensors together with neural networks [13].

Furthermore, for quantified measurements, the system needs to be calibrated for absolute intensities, for which synchrotron radiation can be applied, if available, or a set of several secondary standard sources need to be combined. In both cases, large efforts have to be taken and the calibration is typically only valid for the specific situation during calibration. Even for the alternative (possibly portable) solutions, absolute calibration is mostly not straightforward or relies on calculations. These drawbacks of quantitative VUV spectroscopy lead to scarce investigations of VUV fluxes in plasmas relevant for surface treatment [1,5–7].

In this paper, a solution based on a VUV-sensitive photodiode and interference and edge filters for wavelength selection is presented [14,15]. The portable device is calibrated in-house against an intensity calibrated VUV spectrometer (calibrated down to 46 nm [16]) with the aim to measure absolute VUV emissivities and fluxes flexibly at any low-temperature plasma. Optical filters are chosen according to the most relevant emission systems of a variety of gases and gas mixtures, focusing on process plasmas. For this, absolutely calibrated spectra of inductively coupled plasmas (ICP) from the VUV to the NIR range‡ are analyzed and compared to the occurring ion fluxes (employing a Langmuir probe and an ion mass spectrometer). For the equipped VUV diode system, a thorough characterization and benchmark with the spectrometer is performed and an outlook for general application is given.

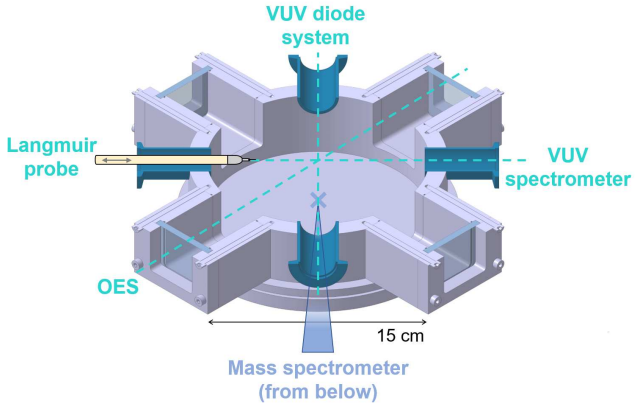

Figure 1. Cut through the laboratory experiment PlanICE, showing the arrangement of the diagnostics (OES: optical emission spectroscopy).

2. Experiment facility

Filter selection, calibration and characterization of the VUV diode system is performed at the experiment PlanICE.

2.1. Laboratory experiment PlanICE

PlanICE is a planar inductively coupled plasma experiment [17]. The cylindrical stainless steel vessel has 15 cm in diameter and 10 cm in height. The planar solenoid is on top of the vessel in an additional vacuum region, separated from the main vessel by a thin quartz plate (3 mm). The vessel is evacuated by a rotary vane pump and a turbo-molecular pump (background pressure some 10^{-4} Pa) and gas supply is provided by mass flow controllers. Plasmas are produced at 2 MHz with maximal 2 kW of RF power, which is sufficient to sustain discharges in the pressure range between 0.3 and 10 Pa for a variety of different gases and gas mixtures. The vessel has various ports for diagnostics, an overview of which is shown in figure 1.

‡ Spectra typical for each gas (mixture) are compiled in the supplemental material, see [link].

2.2. Diagnostics

Diagnostics are available for photon and ion fluxes using an optical and a VUV spectrometer, and a Langmuir probe and an ion mass spectrometer, respectively. The arrangement of the diagnostics can be seen in figure 1.

2.2.1. Photon flux determination For optical emission spectroscopy (OES), radiation is collected via an optic head with a parabolic mirror and transferred via fiber optics to an *Acton SP2750* spectrometer (focal length 0.75 m, grating with 1800 lines/mm). It is intensity calibrated in the wavelength range 200 to 933 nm using an Ulbricht sphere and a deuterium arc lamp. The apparatus profile is Gaussian with a FWHM of 16–22 pm, decreasing with wavelength.

The VUV spectrometer is a *McPherson Model 225* in normal incidence configuration. The focus length is 1 m, the spherical grating has 1200 lines/mm. The apparatus profile is Gaussian with a FWHM of 30–37 pm. Two different detectors can be applied: a solar-blind photo multiplier tube (PMT) for the wavelength range 104.5–300 nm, and a channel electron multiplier (CEM) for the range 30–180 nm. Using a complex in-house calibration procedure and applying five standard sources (see [16]), the spectrometer is intensity calibrated between 46 and 300 nm, where the CEM is used up to 116.5 nm and the PMT above. The spectrometer is connected to the vacuum system of PlanICE via two tube apertures of 4 mm diameter and 7 and 20 cm length, and pumped differentially. The spectrometer can thus be kept at a vacuum of below 10^{-3} Pa even if plasmas with up to 10 Pa are driven in PlanICE. The tube apertures furthermore act as solid angle limiting devices: the accessible solid angle is about 4.8×10^{-5} sr. The corresponding viewing cone of the VUV spectrometer within the PlanICE setup has a diameter of 4.3 mm at the front of the plasma volume increasing to 5.5 mm at the opposite vessel wall.

Using the two spectrometers with three different detectors, spectrally resolved and intensity calibrated measurements in a continuous wavelength range of 46 to 933 nm are enabled, giving access to photon energies between 1.3 and 27 eV. Emission spectra can be given in terms of spectral emissivity or are converted to photon fluxes by integrating over a specific wavelength interval and multiplying by the volume-to-surface ratio of the plasma, 2.1 cm for PlanICE. The thus determined photon fluxes represent the mean value within the detection cone and are assumed to be homogenous for the entire plasma volume.

2.2.2. Ion flux determination To determine the ion flux, the Bohm flux as well as the ion composition is required. The former can be derived from the electron density and temperature from Langmuir probe measurements ($\Gamma_{\mathrm{ion}} \propto n_{\mathrm{e}}\sqrt{T_{\mathrm{e}}/m_{\mathrm{ion}}}$), while for the latter an ion mass spectrometer is required, in particular in molecular gases and/or gas mixtures.

The Langmuir probe is movable, while measurements for the current studies are performed in the center of the vessel (radially and axially). It consists of coaxial conductors, shielded from the plasma by a ceramic tube. The inner conductor is connected to the probe tip, the outer to a compensation electrode used for passive RF compensation at the driving frequency of 2 MHz [18]. Various probe tips have been used, ranging from 50 to 300 μm with lengths of 1–5 mm and made of tungsten or platinum (for pure oxygen plasmas) depending on the discharge gas and the expected electron density. With this approach, the drawn probe current could be maintained within the same order of magnitude despite the extensive parameter variations of feed-gas, pressure and RF power. Together with the RF compensation, distortion of the probe characteristic is thus kept minimal. The electronics as well as the evaluation software are described in detail in [18]. Here, the electron temperature is determined from the second derivative of the probe characteristic and the electron density is determined from the probe current at plasma potential.

An ion mass spectrometer is applied for the determination of the ion composition. The quadrupole mass spectrometer *Balzers PPM 421 plasma monitor* is installed in the bottom plate of the plasma vessel. Ions are extracted from the plasma through an aperture of 100 μm, located 3 cm above the bottom plate, are passed through an energy and a mass filter, and are detected by a secondary electron multiplier (SEM). The system is calibrated for mass-dependent transmission and the ion composition is determined from the energy-integrated mass spectrum.

3. Diode system for VUV flux quantification

In contrast to the VUV spectrometer, the flexible VUV measurement device is developed [14, 15] with the aim of being portable, including its gas-specific calibration.

3.1. Setup

The basic design of the device is shown in figure 2. It is based on a silicon photodiode (*Opto Diode AXUV100G*) that is sensitive between 10 and 1100 nm. A certain spectral resolution can be provided by optical filters mounted on a manually operated filter wheel. It can house five filters, while one of the positions is permanently occupied by a metallic shutter for dark current measurements and to shield the device from radiation during times it is not in use. The set of four filters is then chosen according to the emission

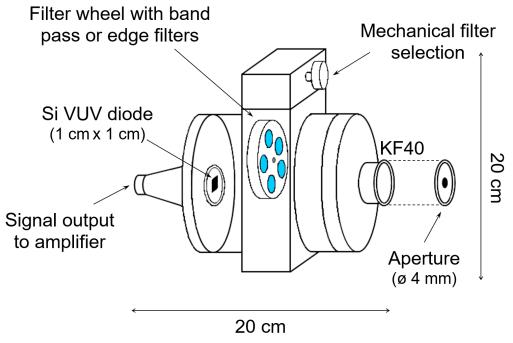

Figure 2. Flexible and portable diode system for measuring VUV radiation.

ranges of the discharge gas under observation (see the following section). The signal from the diode is amplified (*McPherson Model 671*), transferred to a digital multimeter (*PREMA DMM 5017*) and read out by a *Labview* program. The entire device is vacuum tight (electrical feedthrough for the diode signal, mechanical feedthrough for the filter wheel) and is connected to the setup under investigation via an KF flange, without the need for a separate pumping system. An aperture stop of 4 mm diameter within the connection flange limits the collecting solid angle of the device to about 3×10^{-2} sr. The viewing cone diameter within the PlanICE plasma is thus 20 mm at the front and 50 mm at the opposite vessel wall.

The specific viewing cone needs to be considered when comparing measurements between the diode system and the VUV spectrometer or when the diode system is applied at another setup. The former case is of particular importance during the calibration procedure, for which plasmas need to be used that are as homogenous as possible (see section 3.3). Furthermore, the calibration is performed under the specific mutual geometry of the plasma and the diode's detection cone, which define the so-called effective volume. Hence, when transferring the device, a variation of the mutual geometry needs to be taken into account for quantitative measurements (see section 3.4).

3.2. Filter selection

In order to select a set of optical filters for wavelength resolved measurements with the VUV diode system, typically applied feed gases and gas mixtures for process plasmas were analyzed with the VUV spectrometer in a wide range of pressure and power. Discharges in argon, hydrogen, nitrogen, and oxygen together with the gas mixtures hydrogen/nitrogen (50:50), hydrogen/oxygen (85:15), and nitrogen/oxygen (80:20) were studied in the pressure range of 0.3 to 10 Pa. The applied RF power range was individually adapted for each gas and mixture, determined by the sustainability of the plasma at the lower end and the heat load to the components at the upper end. The applied power ranged from 200 W in argon to 1100 W in H_2.

Figure 3 shows an exemplary spectrum between 50 and 900 nm for the nitrogen-oxygen mixture representing synthetic air, i.e. N_2:O_2 80:20, obtained at 1 Pa and 500 W RF power. Prominent emission features are highlighted: below 100 nm several atomic and ionic lines of N and O emit, followed by the resonant atomic lines of N (113 nm and 120 nm, lowest lying transitions in the N quartet system) and O (130 nm, likewise in the O triplet system), the prominent N lines at 149 and 174 nm ending in the metastable states (likewise in the N doublet system), the second positive system of N_2 (SPS, mainly 290–450 nm), the first positive system of N_2 (FPS, mainly from 550 nm upwards) together with further atomic lines of N, and the prominent atomic lines of O at 777 nm and 844 nm. The Lyman-Birge-Hopfield system of N_2 (LBH, mainly 100–200 nm) and emission from the NO molecule, which is formed during the discharge, (γ system, 200–275 nm), can barely be seen in the overview spectrum and are shown enlarged below. The top axis of the overview spectrum gives the corresponding photon energy.

Photon fluxes determined from the spectra are shown in figure 4 for a pressure scan, subdivided into several energy ranges constituting specific emission systems: the N_2 FPS (incl. N and O lines), the N_2 SPS, the NO γ system, an integral between 100 and 200 nm including the N_2 LBH system as well as the atomic resonance lines of N and O, and the integral for the lines below 100 nm with the maximum photon energy of 19.4 eV. The sum of the UV and VUV fluxes (2.6–19.4 eV) is given as well and compared to the occurring total ion flux. Here it is to keep in mind that the ion flux is determined in the center of the discharge, while the photon fluxes are averaged over the detection cone of the spectrometers; the comparison thus represents a lower limit for the photon-to-ion ratio.

It can be seen that the combined photon flux with energies above 2.6 eV is comparable or even larger (in the mid pressure range) than the ion flux for the entire considered pressure range. Only the low-energy photon flux from the FPS surpasses the UV/VUV and ion fluxes, especially for high pressures. Considering the energy resolution within the UV/VUV range, fluxes emitted by the SPS and the 100–200 nm range contribute the most with a comparable magnitude and a maximum between 1 and 3 Pa. From 3 to 5 Pa the flux from the SPS alone is already comparable to the total ion flux. The range below 100 nm is at least a factor of five less intense. But its high photon energy

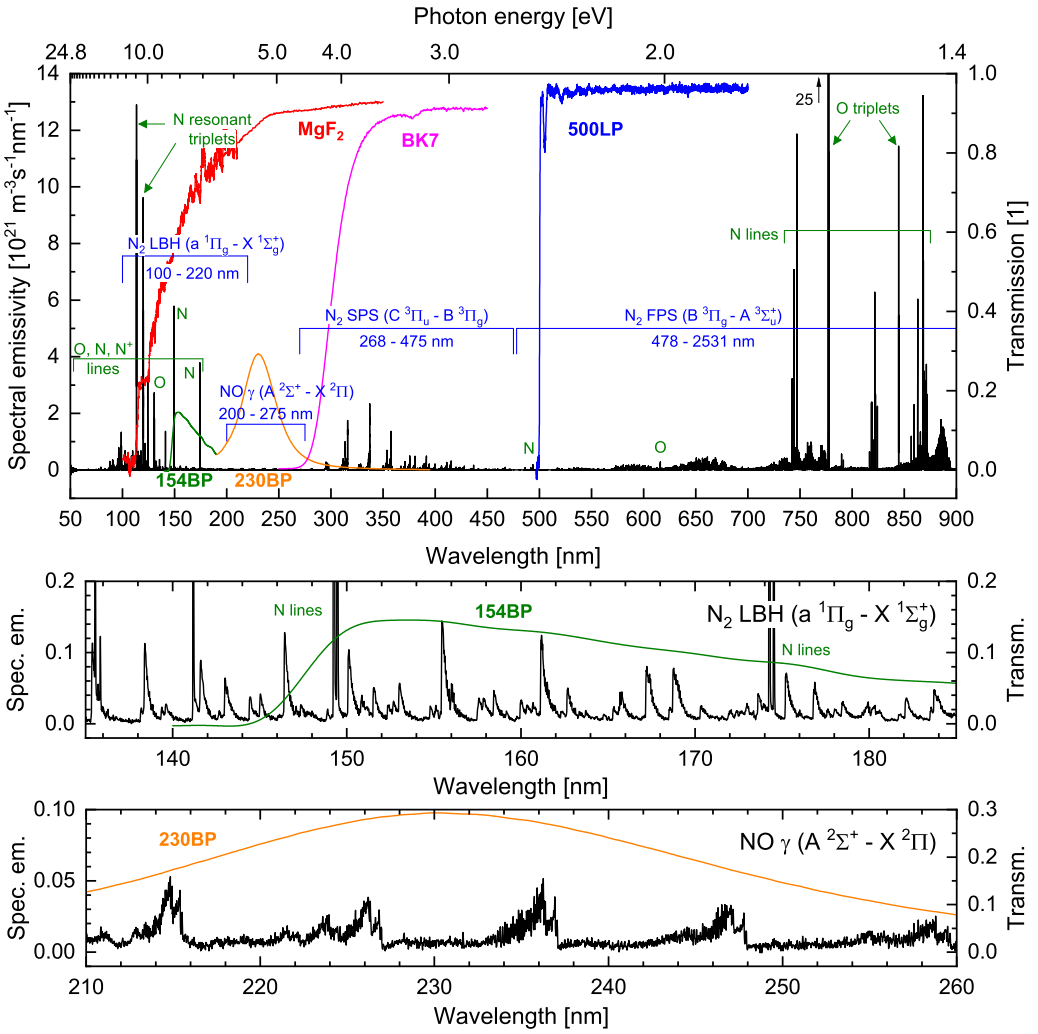

Figure 3. Spectrum of a discharge in synthetic air (N_2:O_2 80:20) at 1 Pa and 500 W RF power. Prominent emission systems are labeled and the transmission curves of five corresponding filters for the VUV diode system are shown. The N_2 Lyman-Birge-Hopfield (LBH) system and the NO γ system are shown separately. Similar spectra for other gases can be found in the supplemental material (see [link]).

close to 20 eV needs to be considered, and fluxes of almost 10 % of the ion flux should not be disregarded in possible plasma-surface interactions. Increasing the pressure, the high energy photons lose relevance and account for only 1 % of the UV/VUV or ion flux at 10 Pa. The γ system of NO, however, clearly gains relevance with pressure, having comparable fluxes with the 100–200 nm range at 10 Pa, thus reaching 25 % of the ion flux.

The varying energetic composition of the photon flux with pressure can also be seen at the mean photon energy that determines the total UV/VUV photon flux: It decreases slightly from about 8 eV at 0.4 Pa down to about 5.5 eV at 10 Pa, which is attributed to the decreasing electron temperature and density with pressure ($9\,\mathrm{eV} \rightarrow 3\,\mathrm{eV}$, $8\times10^{16} \rightarrow 4\times10^{16}\,\mathrm{m^{-3}}$).

A similar analysis is performed for each gas (mixture) [15]. Besides confirming that energetic photon fluxes can reach magnitudes of the ion flux in each of the investigated discharge gases or mixtures, the necessity for an energy resolution of VUV flux quantification is emphasized, since the distribution among the involved emission systems changes with plasma parameters. Hence, appropriate optical filters for an energy resolution of the versatile VUV detection system have been selected. The set consists of five band pass and five edge filters, given in table 1. The actual spectral transmission has been measured in-house with the VUV spectrometer for each filter using discharges in PlanICE as light sources. The transmission curves for the band pass filters 154BP and 230BP as well as for the long pass filters MgF_2, BK7, and 500LP are shown in the spectrum of figure 3. In order to detect specific emission systems or photonic energy

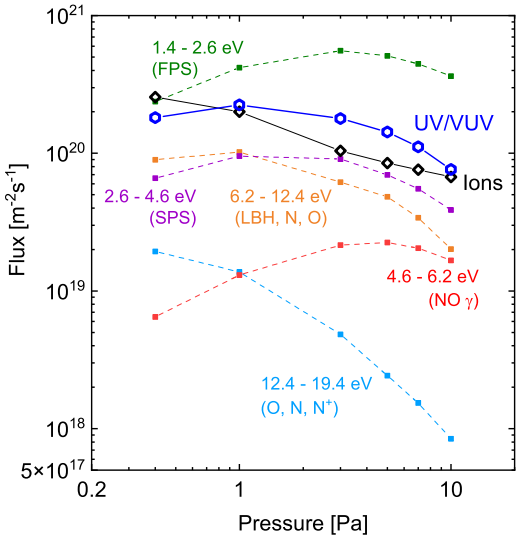

Figure 4. Photon and ion fluxes of a discharge in synthetic air (N_2:O_2 80:20) at 500 W RF power, determined with the VUV spectrometer and the Langmuir probe together with the ion mass spectrometer, respectively. "UV/VUV" denotes the sum over the photon fluxes with energies larger than 2.6 eV.

ranges, either single band pass filters can be used or a combination of two edge filters. For synthetic air, the correlated emission systems are as follows: the MgF_2 window can be used subtractively (i. e. subtracting a measurement with window from one without, labeled hereafter with `1−MgF2` or comparable for the other edge filters) to record the O, N, and N^+ line emission below about 113 nm, the `154BP` transmits part of the N_2 LBH system as well as the two prominent atomic N lines ending in the metastable states, the `230BP` filter only transmits the NO γ system, and the combination of the MgF_2 window with the BK7 edge filter (labeled `MgF2−BK7`) gives the entire emission of the LBH system together with the resonance line of O and the four lowest lying transitions of N (113 nm, 120 nm, 149 nm, 174 nm). Furthermore, using the two filters BK7 and 500LP (labeled `BK7−500LP`) the SPS in the wavelength range between 281 and 500 nm can be selected. This selection is composed of the recommended selections for the pure gases, given in table 2 for N_2 and in table 3 for O_2, together with the specific filter for the N_2/O_2 mixture from table 4. To complete the selection, the appropriate filter sets for hydrogen are given in table 5 and table 6 shows the recommended filter combinations for certain spectral ranges without connection to a specific gas, which can thus be used in any gas.

Furthermore, a set of exemplary spectra (absolutely calibrated from the VUV to the NIR range) for all of the studied gases and mixtures is assembled, each spectrum given with and without the respective filter transmission curves. They can be found in the supplemental material at [link].

Table 1. Available filters for the VUV diode system, four of which can be applied at a time in the filter wheel. Central wavelengths, FWHM and cut-on wavelengths (> 5 %) are determined with the VUV spectrometer.

Band pass filters

Filter name	Central wavelength	FWHM
122BP	122 nm	14 nm
154BP	154 nm	30 nm
230BP	230 nm	37 nm
313BP	313 nm	10 nm
337BP	337 nm	10 nm

Long pass filters

Filter name	Cut-on wavelength
MgF_2	113 nm
Fused silica (FS)	153 nm
BK7	281 nm
400LP	397 nm
500LP	500 nm

Table 2. Recommended filter combinations for the discharge gas nitrogen. "`A−B`" means subtraction of a measurement with filter A minus one with filter B. "`1−...`" means that no filter is installed. The "filter interval" constitutes the spectral range for a transmission larger than 5 %. "LBH" stands for the Lyman-Birge-Hopfield system.

Emission system	Filter combination	Filter interval
N & N^+ lines	`1−MgF2`	$\leq$ 113 nm $\geq$ 11.0 eV
N lines (ending on metastables), part of N_2 LBH	`154BP`	147–187 nm 8.4–6.6 eV
N_2 LBH (a-X), N lines (incl. resonances)	`MgF2−BK7`	113–281 nm 11.0–8.1 eV
combined	`1−BK7`	$\leq$ 281 nm $\geq$ 4.4 eV
N_2 2nd positive system (C-B)	`BK7−500LP`	281–500 nm 8.1–2.5 eV

Table 3. As in table 2 for argon and oxygen.

Gas	Emission system	Filter combination	Filter interval
Ar	Ar resonances, Ar^+ lines	`1−MgF2`	$\leq$ 113 nm $\geq$ 11.0 eV
O_2	O resonance, O^+ lines	`1−FS`	$\leq$ 153 nm $\geq$ 8.1 eV

3.3. Calibration and characterization

In order to perform quantified measurements with the VUV diode system, it is calibrated against the VUV spectrometer. For this purpose, discharges are recorded simultaneously with both VUV systems. Since the solid angle of the two systems is not identical, i. e. the VUV diode system collects radiation from a much larger plasma volume, discharge parameters were chosen for which the plasma is as homogenous as possible. This is analyzed by varying the OES line of

Table 4. As in table 2 for gas mixtures.

Gas mixture	Emission system	Filter combination	Filter interval
N_2/O_2	NO γ system (A-X)	230BP	196–268 nm 6.3–4.6 eV
H_2/O_2	OH band (A-X)	313BP	308–321 nm 4.0–3.9 eV
N_2/H_2	NH band (A-X)	337BP	330–344 nm 3.8–3.6 eV

Table 5. As in table 2 for hydrogen.

Emission system	Filter combination	Filter interval
H_2 Werner band (C-X), H lines $\geq L_\beta$	1−MgF_2	$\leq$ 113 nm $\geq$ 11.0 eV
H line L_α	122BP	115–129 nm 10.2 eV
H_2 Lyman band (B-X)	154BP	147–187 nm 8.4–6.6 eV
H_2 Continuum (a-b)	230BP	196–268 nm 6.3–4.6 eV
combined	1−BK7	$\leq$ 281 nm $\geq$ 4.4 eV

Table 6. As in table 2 for the spectral ranges VUV and UV without connection to a specific gas.

Spectral range	Filter combination	Filter interval
VUV range	1−FS	$\leq$ 153 nm $\geq$ 8.1 eV
UV range	FS−400LP	153–396 nm 8.1–3.1 eV

sight around the plasma: at pressures of 1 Pa and below and high RF powers, a homogeneity of better than 12 % rotationally as well as vertically within the detection cone of the diode system could be determined, and hence calibration is performed at those parameters. To obtain a calibration value, the VUV spectrum is integrated within the filter interval and divided by the corresponding measured signal from the diode in volts. With this approach, gas and filter specific calibration factors are obtained for Ar, H_2, N_2, and O_2 at 1 Pa and the two highest applied RF powers in each gas for reproducibility reasons; the two factors are averaged afterwards to obtain a final value. For gas mixtures, linear combinations of the calibration factors are used, according to the gas composition, or a separate calibration value was determined for newly formed species (see table 4).

It is to keep in mind that this procedure for calibration is only accurate as long as the relative spectrum transmitted through the individual filter is not strongly changed. In particular for gas mixtures, where emission systems from different gases may occur at comparable wavelengths (see e.g. the H_2 Lyman band and a part of the N_2 LBH, both detected through the 154BP filter) this is not obvious a priori. For the presented parameter space of pressure and power, however, the procedure proved to be valid, as shown by benchmark measurements performed throughout the parameter space [15]. Exemplary benchmarks are demonstrated in figure 5, showing power and pressure dependent emissivity or photon fluxes of discharges measured with the diode system and the VUV spectrometer.

Part (a) shows two comparisons in synthetic air: on the one hand, atomic N lines and a part of the LBH system, selected by applying the band pass filter 154BP, and on the other hand, the NO γ system selected by the band pass filter 230BP (see also the enlarged spectra in figure 3). The transmitted (diode system) or integrated (spectrometer) emissivity corresponds to a photon energy interval of 6.6–8.4 eV and 4.6–6.3 eV, respectively. 154BP was calibrated in pure nitrogen at 1 Pa and 400 & 600 W and application in the gas mixture shows perfect agreement with the VUV spectrometer over the entire pressure and power range. 230BP was calibrated in the nitrogen-oxygen mixture at 1 Pa and 500 & 700 W. Hence, the calibration points are included in the graph for the power variation, but the relative trends of the two VUV systems can still be judged. Very good agreement with the VUV spectrometer could be obtained for the performed power and pressure variations. The largest deviations occur at low power or high pressure, and account to less than 13 %.

Part (b) shows the benchmark in hydrogen using filter combination 1−FS which constitutes the VUV range below 153 nm. In this range, the H_2 Werner band and the atomic Lyman series emit (as well as part of the Lyman band), which represent the molecular and atomic resonance systems. Photon energies of up to 15.5 eV have been detected. In H_2 the 1−FS combination was calibrated at 1 Pa with 900 & 1100 W. Apart from the perfect agreement around the calibration point, the relative behavior for the power variation as well as the absolute values for the pressure variation at 700 W agree very well with the VUV spectrometer. The absolute deviation is less than 15 % for the entire parameter space.

In argon discharges, the resonance lines at 104.8 and 106.7 nm can be observed using the filter combination 1−MgF_2, shown in part (c). The calibration point is at 1 Pa, 300 & 400 W, and a slightly increasing deviation can be seen with increasing pressure up to 64 % at 10 Pa. This is an effect of impurity emission lines emerging with pressure in the transmission edge of the MgF_2 window and will be detailed in section 3.4.

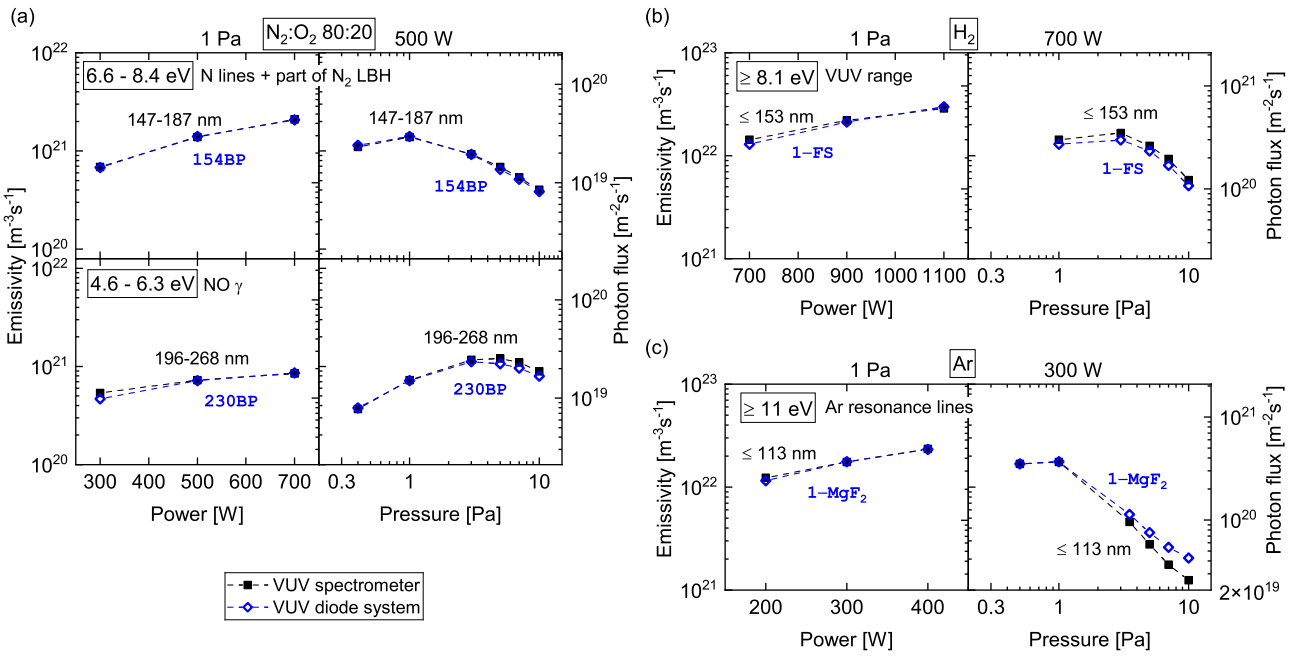

Figure 5. Comparison of emissivities and photon fluxes measured with the VUV spectrometer and the VUV diode system. Pressure and power scans for selected emission ranges for (a) synthetic air, (b) hydrogen, and (c) argon are shown. For the VUV spectrometer the integral range of the spectra is given, while for the VUV diode system the filter combination is denoted. The right axis in each graph is scaled by the volume-to-surface ratio, i. e. it gives the correlated photon flux emitted by the plasma in this spectral range.

With the benchmarked calibration, the VUV diode system is characterized thoroughly for accuracy, stability and reproducibility aspects:

- The benchmark analysis throughout the investigated pressure and power scans of the gases and mixtures showed an overall accuracy compared to the VUV spectrometer of better than 25 %. However, since calibration was performed against the VUV spectrometer, its respective wavelength dependent measurement error needs to be considered additionally. It is given in [16] between 17 % (≈ 123–170 nm) and 60 % (< 116.5 nm) which renders the accuracy for absolutely quantified emission measurements with the diode system wavelength-dependent to 42–85 %.
- The dynamic range of detectable fluxes is more than four orders of magnitude, limited by the signal noise on the lower edge and the saturation of the amplifier on the upper edge. As an example, the accessible photon flux for the Lyman-alpha line in hydrogen is $2.4\times10^{20}\,\mathrm{m^{-2}s^{-1}}$ to $6.1\times10^{24}\,\mathrm{m^{-2}s^{-1}}$.
- A short-term temporal evolution of the measured signal upon irradiation with photons is detected: ± 3 % signal variation can occur within 5–8 minutes of irradiation. While the slope decreases with time, an actual saturation of the signal is not observed. Production of lattice defects (color centers) in dielectric layers of the filter material are suspected. It is thus recommended that the illumination time is limited: in the current studies a maximal exposure time of 30 seconds per filter was chosen.
- The reproducibility of VUV measurements was analyzed during long-term plasma operation. Comparative measurements with both VUV systems were taken every 15 minutes. With increasing plasma-on time, the two VUV systems start to deviate from each other, saturating after about two hours with an offset of the diode system of ≈ + 3 %. Thermal effects are suspected as the cause for this deviation. Letting the entire experimental system heat up for several hours with continuous plasma operation, deviation between the VUV systems is negligible.
- Within three years of operation, and for about 6000 seconds total irradiation time, no evidence for a degradation of the transmission of the filters or the VUV diode could be detected. Hence, it is expected that the system can be used without re-calibration for time scales longer than several hours of total irradiation.

3.4. Application notes

Some aspects need to be considered in experimental application.

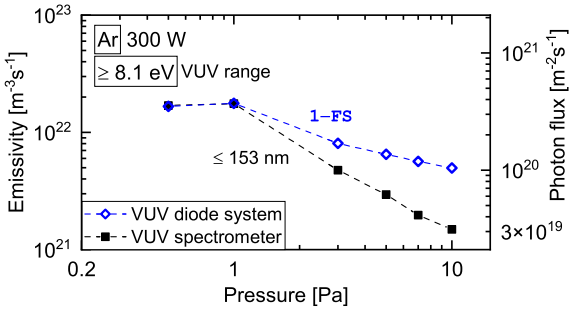

Figure 6. As in figure 5 for argon.

Calibration against the VUV spectrometer is performed by integration of the measured spectrum within the filter transmission interval as defined above. For edge filters, this idealized procedure resembles a step function in contrast to the gradual increase of the actual filter transmission curve. If lines emerge in the edge region of the filter transmission, the correlation of the measured signal with the respective filter range defined in tables 2 to 6 might be disturbed.

Part of this effect is already to be seen in figure 5 (c), but is much more pronounced in figure 6. Here, filter combination `1−FS` is applied in the argon discharge. In the respective wavelength range below 153 nm, the only relevant contribution to the measured signal still comes from the resonance lines. This is confirmed by comparable emissivities or fluxes at low pressure in figures 5 (c) and 6. With increasing pressure, however, the VUV spectra reveal carbon lines emerging in the wavelength region around 156.1 nm. Those lines are still partly transmitted by the fused silica window, even if it is correlated to the range "below 153 nm". Consequently, the diode signal starts to deviate from the integral of the VUV spectrum with 153 nm set upper limit. Emissivity of this line increases by a factor of 40 between 1 and 10 Pa and thus, the flux of the range "below 153 nm" is overestimated by a factor of three. Hence, apart from the distinct definition of filter transmission ranges given above, the real transmission function should be kept in mind.

A similar aspect arises from the lower detection limit of the two systems: the VUV diode measures down to 10 nm while the spectrometer is limited to 46 nm (calibrated range). Hence, emission in the range below 46 nm is not taken into account in the calibration of the diode system but will contribute to the diode system measurement if filter combinations including "no filter" (`1−...`) are used. This might for instance be relevant for discharges containing oxygen, since emission lines of oxygen ions around 43 nm are known.

Another point concerns the rather large detection cone of the diode system, hence it is probing a large plasma volume over which the measured radiation is averaged. This needs to be considered when the plasma under observation emits inhomogeneously, in particular when the results are compared to other diagnostics with differing (smaller) detection volumes (OES, VUV spectrometer). At PlanICE, for instance, pronounced profiles within pure nitrogen discharges emerge with increasing pressure, i. e. plasma emission is more intense closer to the ICP solenoid. Hence, with increasing pressure, the diode system observes a higher intensity than the spectrometer. This leads to deviating results for pressures above 1 Pa.

The last aspect concerns application of the calibration values for measurements at other setups. Since the calibration was performed using low-temperature plasmas in a certain range of pressure and applied RF power, the obtained values are strictly speaking only applicable for plasmas with comparable composition of the emission spectrum. However, as long as the relative spectrum transmitted through the filter is not strongly varied, the calibration values can be seen as accurate (within the given 25 %). This is confirmed by the agreement of diode system and spectrometer for a pressure variation over more than an order of magnitude for a variety of gases and mixtures, by which the relative composition of the transmitted spectra is already varied. Furthermore, even comparing the values of one and the same filter for different gases (e.g. `154BP` for N_2 and H_2), only gives a difference of a factor of two in maximum. Hence, if high accuracy is required also far beyond the parameter space given in section 3.2, the expected emission spectrum certainly needs to be considered. Otherwise, the device can at least be seen as an "order of magnitude" tool with the capability of energy-resolved monitoring of the VUV emission, not necessarily correlated to a specific emission system of the discharge gas.

Furthermore, the variation of the mutual geometry of the plasma and the diode's detection cone compared to the situation during calibration needs to be taken into account. This is done by calculating the so-called effective volume from which radiation is collected: the solid angle of each point within the detection cone spanned by the active area of the VUV diode, is integrated over the volume of the detection cone within the plasma (see [19] for instance). The measured signal is then scaled by the ratio of the effective volume during application to the one during calibration [15]. In case the plasma does not completely fill the observed volume or for complex geometries, ray tracing codes might be required for this purpose (see [20] for instance).

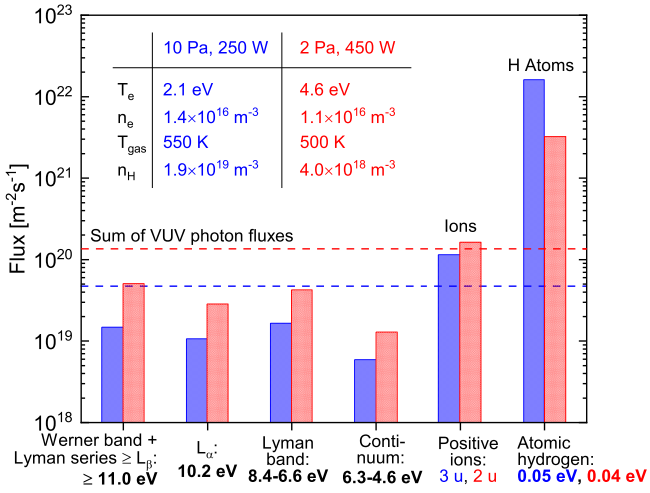

Figure 7. Fluxes measured in hydrogen discharges at the experiment ACCesS: 10 Pa, 250 W in blue, 2 Pa, 450 W in red. The VUV diode system was equipped with the following filter/combinations: `1−MgF2`, `122BP`, `154BP`, and `230BP`. The dashed lines represent the summed photon fluxes. Plasma parameters from Langmuir probe and OES are given for the two conditions; for ions the respective mean ion masses and for hydrogen atoms the respective temperatures (T_{gas} given in eV) are indicated below the bars (color-coded as in the table).

3.5. Application at external setups

As a first application demonstration, the device is transferred in-house to the experiment ACCesS [21]. It is a similar ICP experiment (diameter 15 cm, height 10 cm, RF: 27.1 MHz, max. 600 W) and is typically concerned with investigations on the impact of hydrogen plasma exposure on the surface work function of sample materials [21–24]. Standard diagnostics at the setup include OES and a Langmuir probe, with which fluxes of ions and of the atomic hydrogen radical are determined ($\Gamma_{\mathrm{H}} \propto n_{\mathrm{H}}\sqrt{T_{\mathrm{gas}}}$ with the atomic density n_{H} determined by use of the collisional-radiative model Yacora [25] and the gas temperature T_{gas} determined after [26]). The VUV diode system extends the accessible fluxes to high-energetic photons. Due to the similar geometry of PlanICE and ACCesS, scaling of the calibration factors via solid angle integration is not necessary.

Figure 7 shows a comparison of measured fluxes at 10 Pa, 250 W RF power and 2 Pa, 450 W, typically used for work function investigations. The correlated plasma parameters are given in the table within the graph. VUV fluxes are measured using the following filter/combinations (see table 5): `1−MgF2` for the Werner band together with the Lyman series starting from L_β, `122BP` for the L_α line, `154BP` for the Lyman band and `230BP` for the continuum radiation of the hydrogen molecule.

Emission from the Werner band together with the Lyman series ($\geq L_\beta$) and the Lyman band is thus most intense. Single line emission from L_α is, however, only 30–40 % less intense than the entire molecular systems, highlighting the relevance of the prominent resonance line. The flux coming from the H_2 continuum radiation is a factor of two lower. By decreasing the pressure and simultaneously increasing the power, the electron temperature is increased and the electron density decreases only slightly. The result is an increase of the photon fluxes by a factor of 2–3. The ion flux is only slightly influenced by the parameter variation, while the atomic flux is mainly affected by the decreasing atomic density with decreasing pressure.

For a comparison of particle and photon fluxes, the VUV fluxes are summed and shown as the dashed lines in the figure. At high pressure the ion-to-photon flux ratio is about 2.4, but decreases close to unity at low pressure. In both scenarios, however, the atomic hydrogen flux is dominant by more than an order of magnitude.

As a second application case, the VUV diode system was applied to a high-power large-scale ion source experiment, where VUV fluxes were measured to the very first time [27]. The ion source consists of a cylindrical ICP driver region, where a hydrogen plasma is generated, and a cuboidal expansion region, where the plasma expands and illuminates the plasma grid (PG), the first grid of a multi-grid extraction system for negative hydrogen ions. Measurements have been performed in both regions and the varied geometry of the plasma and the device's detection cone was accounted for: scaling factors were determined to 0.94 and 3.2 for the driver and the expansion region, respectively. The VUV radiation of the Werner and Lyman band together with the Lyman series was measured, giving comparable emissivity for each range and almost two orders of magnitude decreased emission in the expansion region compared to the driver region. The measured emissivities were used to determine the impinging photon flux onto the PG [27]: the main contributing part comes thus from the driver region, while the plasma in front of the PG has only minor influence on the total photon flux. Accompanying measurements with Langmuir probes and OES showed moreover, that ion fluxes are comparable to photon fluxes, while the atomic flux dominates the influx to the PG by more than an order of magnitude.

4. Conclusions

In order to enable VUV flux measurements at low-temperature plasmas, a flexible and easy-to-use device was developed. It is based on a VUV-sensitive photodiode with optical filters for wavelength resolution. The relevant emission ranges of a variety of discharge gases have been determined with the VUV

spectrometer, illustrating the need for energy resolved measurements due to significantly changing spectral compositions of the VUV emission. A set of five edge and five band-pass filters was selected accordingly for the diode system and recommended filter combinations for each discharge gas or mixture are provided, with which specific emission systems can be selected. A compilation of intensity calibrated spectra between 50 and 900 nm with and without filter transmission curves is provided via the supplemental material.

The device is quantitatively calibrated against the VUV spectrometer for absolute flux measurements down to 46 nm. Calibration factors are gas and filter specific with which the device shows an accuracy of better than 25 % for pressure (0.3 to 10 Pa) and power dependent (200 to 1100 W) measurements in a variety of gases and mixtures (Ar, H_2, N_2, O_2, H_2:N_2 (50:50), H_2:O_2 (85:15), N_2:O_2 (80:20)). The validity of the calibration values (or the correlation of filter interval with emission system) outside this pressure range needs to be checked individually, in particular for further gases and mixtures. The dynamic range is more than four orders of magnitude, and the signal stability and reproducibility is better than 3 %.

Portability and applicability of the device is demonstrated by performing VUV measurements at external setups. The determined fluxes could be compared to the occurring ion and radical fluxes, highlighting the relevance of photons due to a VUV-to-ion flux ratio of close to unity.

CRediT authorship contribution statement

R. Friedl: Conceptualization, Methodology, Visualization, Writing – original draft, Funding acquisition

C. Fröhler-Bachus: Methodology, Investigation, Formal analysis, Visualization, Writing – review & editing

U. Fantz: Conceptualization, Supervision, Writing – review & editing

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