論文 / 著書情報
Article / Book Information

題目(和文)	
Title(English)	A Study on Continuum Emission Electron Diagnostics in Atmospheric Pressure Plasma
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出典(和文)	学位:博士(理学), 学位授与機関:東京工業大学, 報告番号:甲第12176号, 授与年月日:2022年9月22日, 学位の種別:課程博士, 審査員:赤塚 洋,小栗 慶之,片渕 竜也,筒井 広明,長谷川 純
Citation(English)	Degree:Doctor (Science), Conferring organization: Tokyo Institute of Technology, Report number:甲第12176号, Conferred date:2022/9/22, Degree Type:Course doctor, Examiner:,,,,
学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	要約
Type(English)	Outline

Dissertation outline

A Study on Continuum Emission Electron Diagnostics in Atmospheric Pressure Plasma

This is an outline of the equally titled doctoral dissertation by Thijs van der Gaag, created at Tokyo Institute of Technology in September 2022. The complete dissertation with relevant references will be published at a later date.

Cold atmospheric-pressure plasmas (CAPP) have been receiving a continuously increasing amount of interest from application researchers for the last two decades. Novel applications have been developed in non-traditional plasma fields such as medical treatments, agriculture and food processing. Besides this, conversion of traditional low-pressure plasma applications to operate at atmospheric pressure is being investigated due to cheaper operation and easier upscaling of atmospheric pressure plasma applications.

Despite the explosive interest in its applications, CAPP has a comparatively short history and the understanding of this plasma behavior is relatively poor. In this thermal non-equilibrium plasma, generally only free electrons have a high temperature of several 10.000 K or several electronvolt, whereas the neutral particle and ion temperature remains near room temperature. This makes electrons responsible for maintaining the plasma and (in)directly producing the desired radicals or reactive species for a CAPP application. As such, understanding the electron population and its energy distribution is most essential.

For the determination of the electron energy distribution function (EEDF) in CAPP, assumption of the commonly used Maxwell distribution [~exp(-E/kT_e)] or Druyvesteyn distribution [~exp(-(E/kT_e)^2)] is not recommended since its validity is not proven. Instead, the arbitrary EEDF should be determined directly from experimental results. For this, a data continuum is required, which can be provided by the V-I curve of an electric probe or the line broadening of free electron scattering in Thomson scattering. However, no relevant theory exists yet for the use of probes in high- and atmospheric-pressure plasma and although Thomson scattering at atmospheric pressure is a highly accurate method both temporally and spatially, it is practically difficult to use and requires high expertise with sophisticated equipment. From this, the need for new electron diagnostics method capable of determining the EEDF is high. In this study, the continuum spectrum measured by optical emission spectroscopy (OES) is investigated for EEDF determination.

The continuum spectrum in CAPP is measurable with conventional OES equipment and is dominated by electron-neutral free-free bremsstrahlung in the visible spectrum. This neutral bremsstrahlung is related to the EEDF through the following incomplete Volterra integral equation:

$$\varepsilon_{ea}(h\nu) = \int_{h\nu}^{\infty} R(h\nu, E) f(E) dE.$$

Here $\varepsilon_{ea}(h\nu)$ is the neutral bremsstrahlung emissivity, $R(h\nu, E)$ is the so-called kernel equation which is a known function of photon energy $(h\nu)$ in electronvolt and electron energy (E) in electronvolt. Finally, $f(E)$ is the electron energy probability function (EEPF), which is related to the EEDF by: EEPF=EEDF/$\sqrt{E}$. This Volterra integral is classified as an incomplete Volterra integral, since practically the electron energy and photon energy dimensions do not have the same size. With conventional OES equipment, the visible spectrum (300-800 nm) can be measured, which corresponds to a 1.5-4 eV photon energy range. The desired EEPF determination range is 0-20 eV electron energy. This makes the information available (continuum spectrum) smaller than the desired target (EEPF). A consequence of this is that no existing methods for complete Volterra integral solving were found that could be applied to this incomplete Volterra integral as well.

A method for inverting the Volterra integral is created, which is based on EEPF simulation and statistical analysis. To accelerate the process, reinforcement learning is applied. The schematic overview of this method is shown in Figure 2.11 and is titled the Visible Bremsstrahlung Inversion (VBI) method. Its inputs are the continuum spectrum from an OES measurement, the solution space, which defines the EEPF simulation limits and numerical discretization of the simulated EEPF population and finally the electron-neutral momentum-transfer cross section. The output is the best arbitrary (numerical) EEPF result, its statistical uncertainty and fitness.

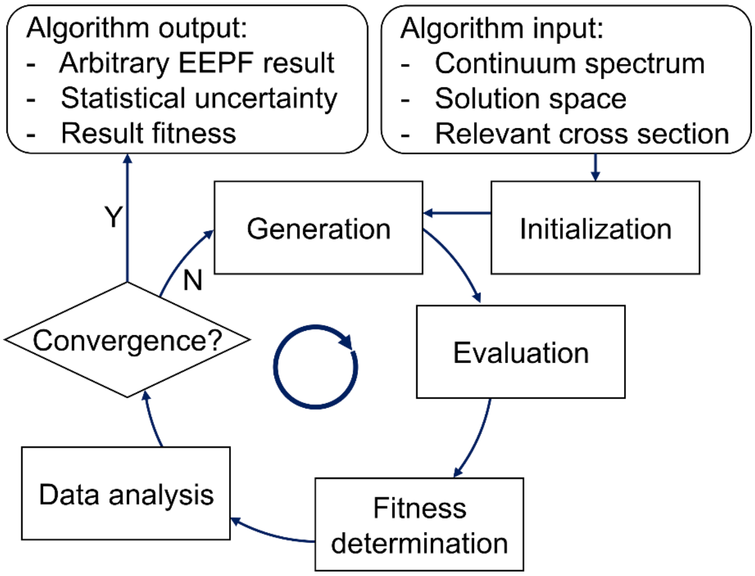

Figure 2.11. Overview of the VBI scheme with its main components.

In this process, numerical EEPF are generated within the defined solution space. This numerical EEPF population is then substituted in the neutral bremsstrahlung integral equation and the integral is numerically evaluated to obtain the calculated emissivity $\varepsilon_{calc}(h\nu)$. The calculated emissivity is then compared to the OES measurement and a fitness is assigned to each individual EEPF. In the data analysis phase, the optimal generation distribution for the next iteration is created following the objective function $|\varepsilon_{ea} - \varepsilon_{calc}| \rightarrow 0 \; \forall \; h\nu$. Through this iterative process, convergence through the true EEPF can be obtained. Theoretical verification results using an assumed Druyvesteyn EEPF can be seen in Figure 2.43. For this, 10^7 generated functions were evaluated over 100 iterations, calculating order 10^9 numerical integrals, which took a total of 6.25 minutes using optimized GPU (RTX 2080Ti) calculation.

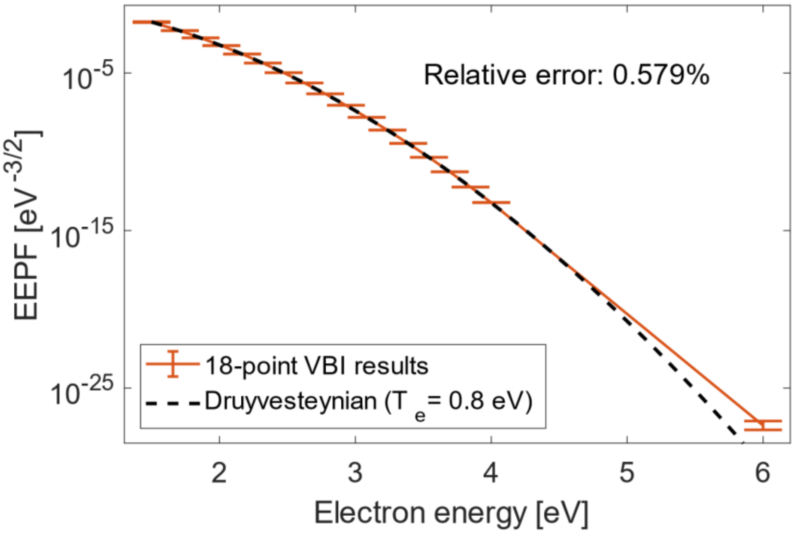

Figure 2.43. Verification results of the Druyvesteyn case. The corresponding spectrum has a mean relative error of $2.19 \cdot 10^{-3}$ with respect to the input spectrum.

A practical challenge to use this method is the isolation of the continuum spectrum. For this, a peak analysis tool was developed that effectively removes the line spectrum from an OES measurement. It can easily be configured to suit any spectrum measurement. The peaks are detected as local maxima with a configurable minimum prominence and maximum peak width. These detected line emission peaks are then replaced with a linear interpolation. Both attachment points and the gradient of this linear interpolation are optimized to minimize the gradient difference between the attachment points and to remove as little as possible of the original spectrum while effectively removing each peak individually. The results of this are shown in Figure 3.23. The full spectrum analysis flow from measurement to isolated continuum spectrum including second-order diffraction filtering and optical calibration is shown in Figure 3.24.

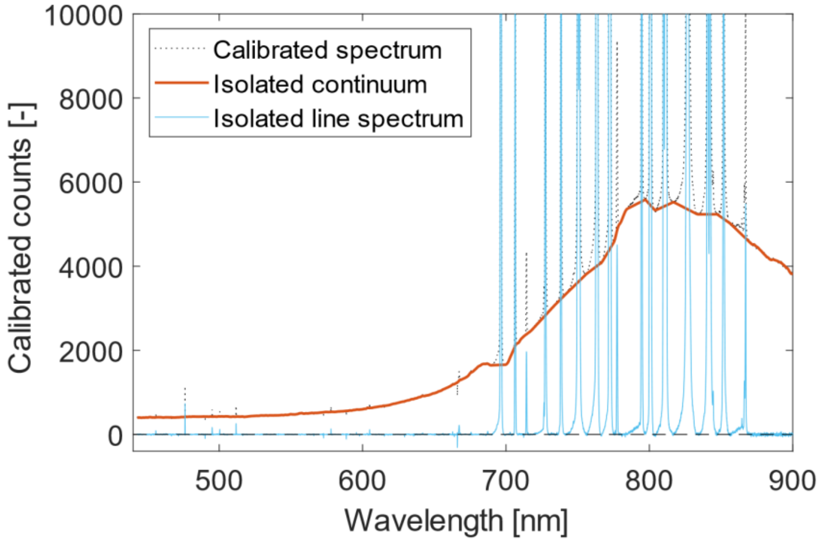

Figure 3.23. The calibrated experimental spectrum with its isolated continuum and line spectrum components

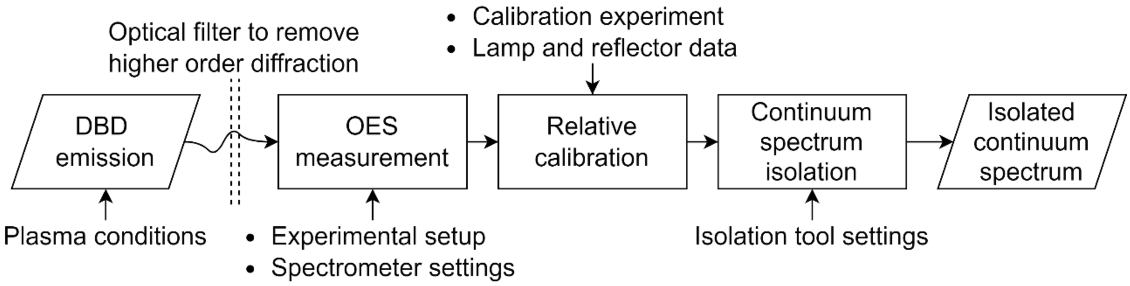

Figure 3.24. Flowchart of the spectrum analysis from emission to isolated continuum spectrum.

Finally, the limitations of the VBI method are discussed. Using OES and a statistical method means that both can impose limitations on the method. The resolution and electron energy range in which the EEPF can be determined depend on both the quantity and quality of the available information. The quantity of this information is determined by the OES measurement range as is shown in Figure 4.5 in photon energy-electron energy space. An OES measurement from 1.5-4 eV photon energy yields an EEPF with high resolution in the low energy region (1.5-4 eV electron energy). Neutral bremsstrahlung emission in the low energy region is both measurable and highly dependent on the electron energy of the electron responsible for that emission. This allows determination of the EEPF with high resolution. Below 1.5 eV, no information is available and the EEPF cannot be determined. Furthermore, the EEPF can be determined in the high energy region with low resolution. The neutral bremsstrahlung emission from the high energy region is measurable by OES, but electrons in this region can emit photons anywhere in the spectrum. This makes the information from these electrons more difficult to access, leading to a generally low EEPF resolution. This effect can be seen in Figure 2.43, where a theoretical spectrum was calculated from 1.5-4 eV photon energy. The verification results show a high resolution in the 1.5-4 eV electron energy, while only one point is placed in the high energy region.

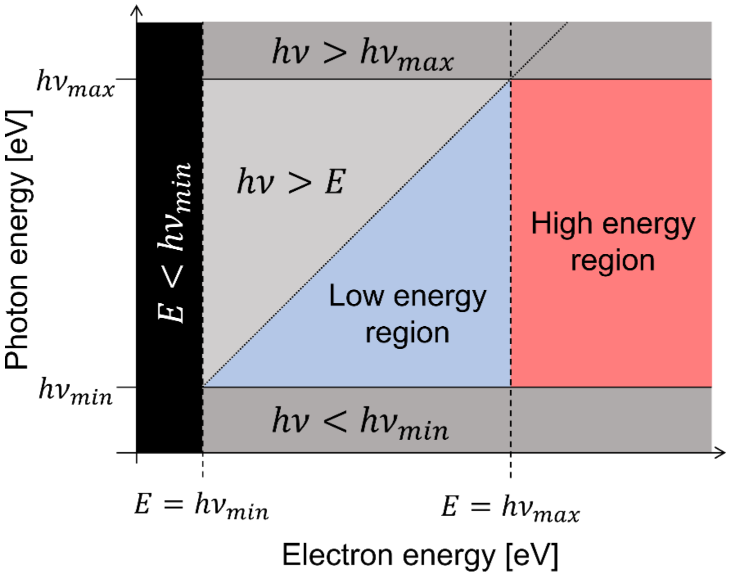

Figure 4.5. An overview of the limitations of the VBI method in different regions in the electron energy-photon energy space.

The quality of the obtained information depends on whether the significant portion of the measured neutral bremsstrahlung spectrum originates from the low or high energy region. In figures 4.2 and 4.3 the kernel function multiplied with a T_e = 1 eV and 2 eV Druyvesteyn EEPFs are shown in photon energy-electron energy space. The red area shows the highest intensity neutral bremsstrahlung, whereas the dark blue area indicates the space with no significant neutral bremsstrahlung emission. It can be clearly seen that this area shifts from the low energy region to the high energy region with increasing electron temperature. Based on this, it is assumed that this method can be used mostly for $T_e < 2$ eV with the limitations as described above. For an electron temperature beyond 2 eV, this method is still usable, but the limitations will change. Generally, CAPP has a low electron temperature. It is expected that this method can be applied as shown.

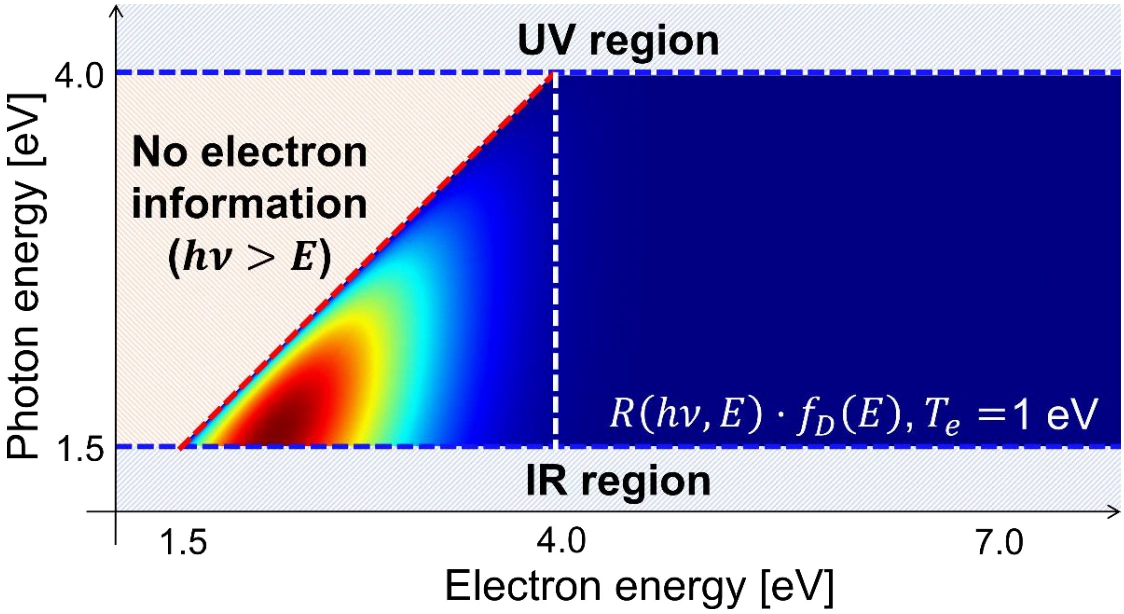

Figure 4.2. Argon *Rf* overview using a Druyvesteyn EEPF with T_e = 1 eV.

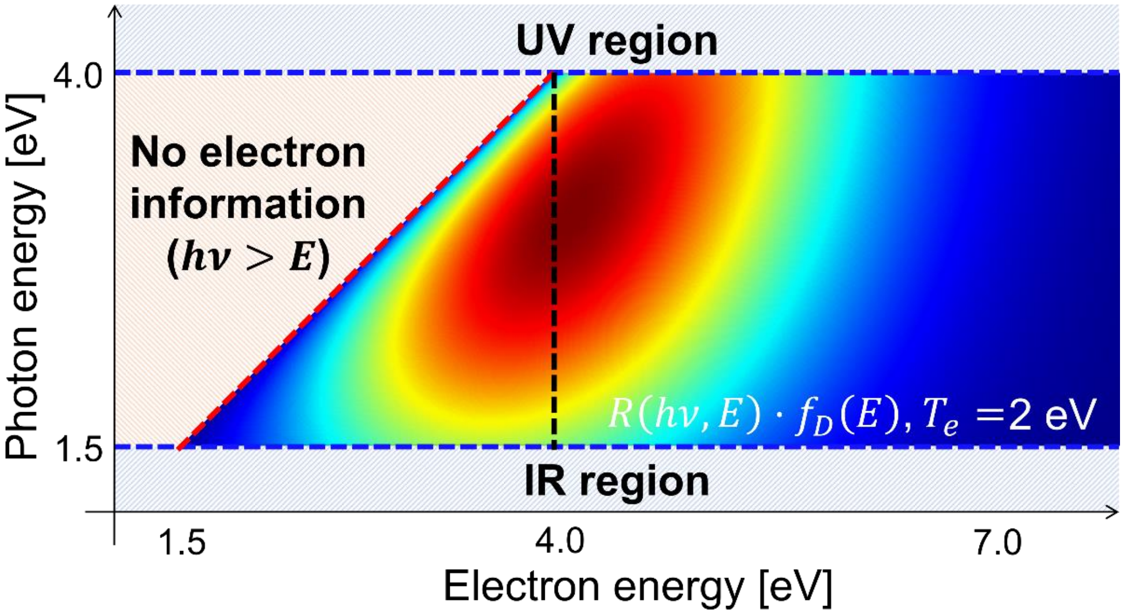

Figure 4.3. Argon *Rf* overview using a Druyvesteyn EEPF with T_e = 2 eV.

In conclusion, a new electron diagnostics method was created that is capable of determining the arbitrary or numerical EEPF in CAPP. It is a highly accessible method, requiring only basic OES equipment with relative or absolute intensity calibration and the well-established electron-neutral momentum-transfer cross section, which can be obtained from databases such as LXCat. It was shown that the VBI method can successfully obtain a partial EEPF with high resolution and low uncertainty through machine-learning-accelerated statistical analysis. A continuum spectrum isolation pipeline was shown with all steps required to obtain a clean continuum spectrum from an OES measurement. For this, a peak analysis tool was developed that is capable of removing the line spectrum from an OES measurement. This tool does not rely on any line emission databases and is a general tool that can be used for any spectrum. Finally, the limitations of the VBI method are discussed. In summary, a 1.5-4 eV photon energy visible range spectrum (300-800 nm) yields an EEPF with high resolution from 1.5-4 eV and low resolution in the 4-20 eV range. The upper limit of 20 eV was established through a sensitivity study and might change depending on the plasma source.

Easily accessible EEPF measurement methods are highly desired for CAPP. This method provides access to a partial EEPF from simple OES equipment and a single cross section. It is desired that this method is rapidly integrated in the CAPP community for electron diagnostics as well as further improvements of the VBI method.