Experimental transition probability measurements in pulsed lamps: critical points

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Abstract. Pulsed linear lamps may be very useful experimental devices to obtain transition probability data. They can provide valuable information of the spectral emission of spectral lines in very different conditions of electron density and temperature. However, they require a very careful previous analysis, which include many experimental aspects e.g., spatial and temporal emission homogeneity, good characterization of equilibrium status or a very precise knowledge of the equipment employed to make measurements: spectrometer transmittance, self-absorption control or lamp windows dirtying. Most of these aspects will be considered along this work, their particular relevance as well as the way they have been controlled in the Plasma Physics Laboratory at the University of Valladolid (Spain).

1. Introduction

The importance of transition probabilities (A_{ki}-values) is well established. In many scientific, industrial or astrophysical fields, transition probabilities are used to determine plasma temperature and, from this, element abundances and other physical parameters of interest can also be obtained. Pulsed lamps may be very promising sources of high quality/low uncertainty transition probability data. Among its intrinsic advantages, it is remarkable the possibility of making measurements at very different plasma conditions. However, several critical points should be very carefully considered when designing and using these light sources for this scientific purpose.

The calculation of the geometry involved in the process emission-detection of the radiation coming from a spectral line makes commonly the determination of its absolute transition probability an unfeasible task. In this case, only relative transition probabilities are available. However, when only one relative scale is tried and lines coming from different upper levels are involved, the characterization of the equilibrium state in the plasma becomes a new significant problem. In this case, the existence of excitation equilibrium along the measured plasma volume should be carefully guaranteed. The Boltzmann-plot technique is a useful tool to check the exponential population of the excited states of the radiation emitters. An absolute scale may be again attainable if a reliable set of
transition probabilities exists previously in the literature and is used to perform the Boltzmann-plot and therefore determine the population temperature.

The experimental difficulties are not negligible either. If the uncertainty associated to the determined transition probability value is tried to be low, it is necessary a high homogeneity in the emitting plasma volume whose radiation is detected. This requires, among others, the absence of cold boundary layers which may emit the same spectral lines at very different intensities than those emitted in the hottest regions. Furthermore, although the homogeneity might be checked in a reliable way, this is not enough. Self-absorption may become a serious problem whose control is of vital importance. When the quantification of this effect is possible and is not too severe, different experimental and numerical procedures exist in the literature which may help us to reconstruct the original emitted profile and to make possible the comparison of line intensities.

From the experimental point of view, other cautions should also be considered and its effects quantitatively evaluated. Among them, we could remark the need of a precise calibration of the spectrometric system and the possible influence of electrode sputtering on spectral transmittance of lamp windows.

In this work, we will face most of the critical points that should be considered for a precise determination of the $A_{ki}$-values as well as the strategies employed in the Plasma Laboratory at the University of Valladolid.

2. Experimental considerations

From now on, we will be concerned about plasma spectroscopy emission experiments where the plasma is generated by discharging a capacitor bank, charged up to 7000-10000 V, through a cylindrical Pyrex tube. The required characteristics of the electrical discharge employed in our laboratory are the following: a) pulse width in the range 100-300 $\mu$s, b) peak current in the order of kA with an energy near kJ to produce dense plasmas, c) smooth current variations with time, which prevent the plasma shock waves and so, of instabilities. d) about a 40 $\mu$s plateau, that gives measuring facilities and, finally, a high reproducibility in the pulses.

The excitation unit has been designed, constructed and tested in our laboratory. Further details can be found elsewhere [1]. The capacitors are connected to one another and to the discharge system (a Spark-Gap is used) by self-inductions which reduce the temporal derivative of the intensity of the current. The self-induction values are selected in order to obtain a current pulse with a stable 40 $\mu$s plateau at the first discharge instants, followed by a downfall close to critical damping, which actually shows a small plateau between the instants 90-110 $\mu$s (see Figure 2 in [2]). In order to guarantee the best plasma repetitiveness the excitation unit has a continuous current source of low intensity (several mA) that allows working with a preionised gas in the lamp.

In its basic design, the plasma source consists of a cylindrical tube of Pyrex glass, 155 mm in length with an inner diameter of 18 mm. The figure 3 in [2] shows a section of the lamp in which all the relevant data have been specified. The glass tube is projected farther than the electrodes, made of aluminum, to reduce sputtering. The windows, which enclose the lamp, are separated 10 mm from the electrodes by a metal ring, electrically isolated by means of viton o-rings from the electrodes. In this configuration the distance between windows is 175 mm. This basic design allows several thousands of discharges to be performed without significant damage on the window surfaces. The electrical connections to the electrodes are made using 4 rigid rods on each electrode, parallel to the lamp axis and symmetrically distributed around it, to avoid plasma inhomogeneities during the discharge. The lamp is connected to the vacuum and gas systems by standard connections placed on the electrodes.

The gas or mixture of gases are flowing continuously through the tube at pressures of several hundreds or thousands Pa. An example of the typical experimental arrangement has been previously published [3]. In this arrangement the lamp is placed in one of the arms of a Twyman-Green interferometer. Two laser beams (543.0 nm and 632.8 nm) pass through the lamp in the axial direction in order to obtain the temporal evolution of plasma refractivity and from this, the electron density ($N_e$) with an uncertainty lower than 10%. Typical highest values in $N_e$ are around $2\times10^{23}$ m$^{-3}$. Spectroscopic
and interferometric end-on measurements are usually performed simultaneously during the plasma life, both at 2 mm off the lamp axis in a symmetrical configuration. The spectroscopic beam is limited by two diaphragms 2-3 mm in diameter, and focused by a concave mirror of 150 mm focal length onto the entrance slit (35-70 μm width) of a Jobin-Yvon monochromator (1.5 m focal length, 2400 lines mm\(^{-1}\) holographic grating). The spectra are currently recorded with either an optical multichannel analyser (OMA) of 512 channels (EG&G1455R-512HQ) or an intensified charge couple device ICCD (4QuickE Stanford Computer Optics). Measurements are usually performed in first order of diffraction with an exposure time around 1-5 μs. Excitation temperature is determined by a Boltzmann-plot of the species of interest and is usually in the range 15000–35000 K. The error in temperature measurements is estimated to be around 10%.

3. Spatial and temporal homogeneity of the discharge

One of the key points which should be considered when measuring transition probabilities from the line emission of spectral lines is that the plasma parameters should be reasonably homogeneous in the measured plasma volume during the exposure times of the detection system.

Concerning the temporal homogeneity, only the instants where temperature and electron density show the lowest gradients should be selected. In relation to the spatial homogeneity, the situation is much more complex and very often only indirect information can be obtained from the experiment after a very laborious analysis.

Figure 1. Scheme of the lamp employed for Stark parameters and transition probability measurements of Si II and Si III spectral lines. Protective chambers 30 mm long were employed in both extremes of the lamp.

An example of the analysis of the inhomogeneities along the plasma column was performed for a specific design of lamp built for measuring Stark parameters and transition probabilities of Si II and Si III [4,5]. The plasma was generated on a mixture of silane (SiH\(_4\)) and helium. In Fig 1 this design is shown. To the basic design described above, protective chambers were added to both extremes of the lamp. Furthermore, entrance and exit of gas in the lamp is made from these two chambers. This technique reduces very efficiently the lamp windows dirtying by sputtering and the deposition of sand on the extremes of the lamp by creating a laminar flow of gas just on the inner side of windows surfaces. Chambers of different lengths were tested by comparing the electron density evolution curves obtained from interferometry and from Stark broadening of some He I lines. Results are shown in Fig. 2. In the determination of the interferometric electron density curve, it has been assumed that the plasma column length is equal to the lamp length.
As concluded from Fig. 2, the very good agreement obtained between both determinations of $N_e$ when no chambers are included (basic design) or with chambers 30 mm length in the specific design, informs us that the plasma column fills completely the lamp under these two designs and thus, if cold boundary layers exist, they do not affect significantly to the electron density determination. For longer chambers, the agreement is much worse and it might be reasonably to conclude that the plasma column length changes during the discharge.

Additional evidences of the plasma homogeneity can be obtained with some more experimental work. In previous works [2], the results obtained by axially measuring the interferometric electron density evolution curve at different distances from the lamp axis were published. A similar analysis was performed for the He I excitation temperature evolution curve obtained from Boltzmann-plot. From these experimental results we conclude that, after some tens of microseconds, there exists a highly homogenous and symmetric core within the cylindrical plasma column. This would allow, for instance, to work with light beams of 2-3 mm diameter in the plasma column for spectroscopic and interferometric standard measurements without important loss of spatial resolution. This symmetry also allows to perform interferometric and spectroscopic measurements in separated but symmetrical plasma column relative to the lamp axis. Furthermore, the temporal distribution of the main plasma parameters with their soft evolution allows good spectroscopic registers within the time scale of pulsed plasmas, and along a wide range of different values.

An additional study which would inform us about the spatial resolution of the discharge is the analysis of the symmetry of theoretically symmetric profiles, as, for instance, some hydrogenic lines. This analysis was performed by us for the He II $P_a$ line [6]. Results revealed levels of profile symmetry never published before in the literature.

![Figure 2. Comparison of the interferometric and spectroscopic determinations of $N_e$ for three different chamber lengths and for our basic design (no chamber).](image)
4. Plasma equilibrium mode

The level of knowledge of the plasma equilibrium state depends, among others, on the procedure employed for obtaining the $A_{ki}$-values. If we desire to obtain the transition probabilities of lines coming from the same upper energy level in an emission spectroscopy experiment, no equilibrium specifications are required. Only the comparison between intensities of the involved lines will provide the transition probabilities in a relative scale, which can be converted in an absolute one if one of them is previously known.

If lines coming from different upper energy levels are pursued in a common scale, the situation becomes more complex. In this case, the minimum equilibrium requirement corresponds to the partial local thermodynamic equilibrium (pLTE). Under these conditions, the upper energy levels are assumed to be populated according to the Boltzmann law. The classical Boltzmann-plot is a good way to check this type of equilibrium. This procedure requires that some transition probabilities are previously known. The presence of a linear behaviour in these plots allows us to define an excitation temperature $T_{exc}$, parameter from which the $A_{ki}$-values can be obtained by simply considering its measured intensity. A possible departure from this linear behaviour would point out to the existence of non balanced atomic interaction processes inside the plasma [7]. However, it is important to remark that, both the analysis of the plot linearity and the final quality of the obtained transition probabilities are conditioned by the average uncertainty of the $A_{ki}$-values taken from the literature. Furthermore, the existence of a linear behaviour in the plot only guarantees the pLTE state for the range of upper energy levels involved, not for higher or lower ones. Transition probabilities obtained, from this $T_{exc}$-value, for lines coming from upper energy levels out of this range should be very carefully considered.

In Fig. 3 some of these plots are performed for different instants of the plasma life in a mixture of xenon and helium, as well as the obtained $T_{exc}$. Only Xe II lines are considered. As shown in this figure, a very good linear dependence is obtained for the instants in which this analysis has been made.

![Figure 3. Boltzmann-plots and $T_{exc}$-values obtained from Xe II lines. Considered times of the plasma life are expressed in microseconds and the correlation factor of the linear fit is included as an index of the level](image)

Although other equilibrium models are possible, they usually require more theoretical assumptions not always easy to be checked experimentally. These assumptions finally derive in greater uncertainties and mistrust on the final transition probabilities.
5. Control of self-absorption

It is frequent that, at the plasma conditions from which the pursued transition probabilities are available, some profiles may show self absorption. It is important to control this parameter if we desire to properly measure the intensity emitted by the line. In order to make this control, it is usual to place a mirror behind the lamp. This mirror allows the radiation leaving lamp in the opposite direction to the detection system, to reenter the lamp and adds to the radiation directly emitted towards the spectrometer. In a certain instant of the plasma life and for each wavelength, the absence or presence of selfabsorption in the measured profile can be checked by simply comparing the light registered from each line when the self absorption mirror (M3 in Fig. 1 of [3]) is covered and when this mirror is not covered. The ratio of the registered intensity in these two spectra, gives us the effective reflection coefficient of the mirror which should be constant if selfabsorption does not exist (see Fig. 4). If self-absorption appears in some part of the spectrum, this reflection coefficient is strongly diminished as happens in Fig. 4 for the Ar II line 493.3 nm. Different reconstruction techniques of the original profiles are available [8]. All them require a high enough spectral transmittance in the lamp window closer to the self absorption mirror.

![Figure 4](image-url)

Figure 4. Original (solid squares) and reconstructed (dotted line) spectra containing some Ar II lines. Reflection coefficient is also plotted by a solid line.

However, the sputtering coming from the electrodes after each discharge produces an embedding of electrode materials in the lamp windows inner surfaces. This produces two annoying effects which should be considered all along the process of transition probability determination. Fist, the sputtering strongly reduces the spectral transmittance of the lamp window closer to the self-absorption mirror. This makes more difficult the self-absorption check and even may make impossible the reconstruction process of selfabsorbed spectra. Second, even although selfabsorption may not exist, the loss of spectral transmittance of the window lamp closer to the spectrometer should be considered to obtain the really emitted intensity of spectral lines by the measured plasma column. In Fig. 5 several curves of spectral transmittance of the lamp windows are shown. The upper one corresponds to a clean window and the following ones below it correspond to the spectral transmittances obtained with an increasing number of discharges. As can be clearly seen, the loss of transparency is dramatic, particularly at the lower wavelengths.
As can be seen in this figure, there are wavelengths ranges where transparency increases in spite of the increasing number of discharges. This is due to the interference effect of a multilayer deposition on a surface, whose spectral transmittance depends on its thickness and wavelength.

![Figure 5](image1.png)

**Figure 5.** Spectral transmittance for different number of discharges performed.

Concerning the spectral transmittance of the detection system, the most significant aspect to be considered is the effect of the spectrometer on the final detected counts per line. Both the monochromator and the detector have a spectral response strongly dependent on wavelength. Furthermore, each channel of an unidimensional or a bidimensional detector array may have different spectral responsivity, which complicates the correction of the detected spectra of the spectral transmittance function.

In Fig. 6 the bidimensional spectral transmittance function (in arbitrary units) of a spectrometer formed by a Jobin-Yvon monochromator, first order of diffraction) and an ICCD (4QuickE Stanford Computer Optics) is shown. This function is obtained by considering the obtained spectra from an incandescent lamp emitting as a blackbody at a colour temperature of 3041 °K from which a previous spectral calibration is obtained. As shown in this figure, the spectrometer employed in this experiment is enhanced for the ultraviolet radiation detection. As mentioned before, the wavelength is the dominant effect on the spectral transmittance function but however, the influence of the detector
channels where a spectra line is registered is very important on the final number of counts registered.
This is a typical effect of the optics involved in the ICCDs.

6. Conclusions
Measurement of transition probabilities in pulsed discharge lamps is a complex experimental task. After the experience compiled during the last 30 years in the Plasma Laboratory at the University of Valladolid, we have learnt that, although theoretical assumptions concerning equilibrium are usually verifiable and not too strong (pLTE), the plasma source experimental set-up, its proper featuring and the control of all the aspects involved in the line radiation measurement are difficult challenges. Very good spatial and temporal homogeneity features are required and much previous experimental work is to be done until a plasma discharge can be considered as valid for this kind of measurements. Other factors, such as selfabsorption, spectral transmittance or the control of windows dirtying should also be very carefully considered if a low uncertainty is aimed in the final $A_{ki}$-values.

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