Transition Probabilities of 30 Pb II Lines of the Spectrum Obtained by Emission of a Laser-Produced Plasma

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Abstract

Transition probabilities have been determined for 30 lines of Pb II by measuring the intensities of the emission lines of a laser-produced plasma (LPP) of Pb in an atmosphere of Ar. The plasma has been seen to contain local thermodynamic equilibrium (LTE) and homogeneity; the plasma studied has a temperature of 11 500 K and an electron density of $10^{16}\,\mathrm{cm}^{-3}$. The experimental results obtained during this study have been compared with the experimental and theoretical values given by other authors.

1. Introduction

It is well-known that a knowledge of transition probabilities is relevant for studies of atomic structures, astrophysics and lasers, as well as for different analytical techniques. In the case of lines of Pb II, very few experimental results are to be found in the specialized literature [1–3] and, of the few that do exist, some differ to quite a large extent.

This has been the reason for two studies, one of which is the Ref. [4], that presents the transition probabilities for 30 lines corresponding to levels S, P, D and F of Pb II, using the emission of a plasma produced in a hollow cathode discharge, and this study, where transition probabilities are obtained for 30 lines corresponding to levels S, P, D and F of Pb II obtained by emission of a plasma generated by focusing a laser beam on a sample of lead. This data includes new values for 10 transitions that it was not possible to study in [4].

The interaction of a high power laser beam with solid samples generates plasmas on the surfaces of the targets with high temperatures and electron densities; this process has drawn considerable attention over recent years as a result of the information that the plasmas provide [5–7].

The corresponding absolute transition probabilities were obtained by plotting the intensities of the Pb II emission spectrum lines on a Boltzmann plot assuming local thermodynamic equilibrium (LTE) to be valid and following Boltzmann's law. The values obtained during this study are compared with existing experimental and theoretical values.

A study has been made of the temperature, electron density and plasma homogeneity, discussing the LTE hypothesis.

2. Experimental setup and procedure

A block diagram of the experimental setup is shown in Fig. 1. The apparatus and methods used are similar to those used in the spectrum study of Na II [8] and Si II [9]. A Q-switched laser Nd : YAG (Quantel YG585) produces 10 ns pulses of 275 mJ at 10 640 Å with a frequency of 20 Hz. A lens with a focal distance of 12 cm is used to focus the laser beam on a lead target having a purity of 99.9%.

A chamber is used to generate the plasma, in a vacuum or in a gas atmosphere; after a vacuum of 10^{-5} Torr had been attained inside the chamber, by means of a turbomolecular pump, it was filled with Argon and maintained at a constant pressure of 6 Torr throughout the measurement, using a

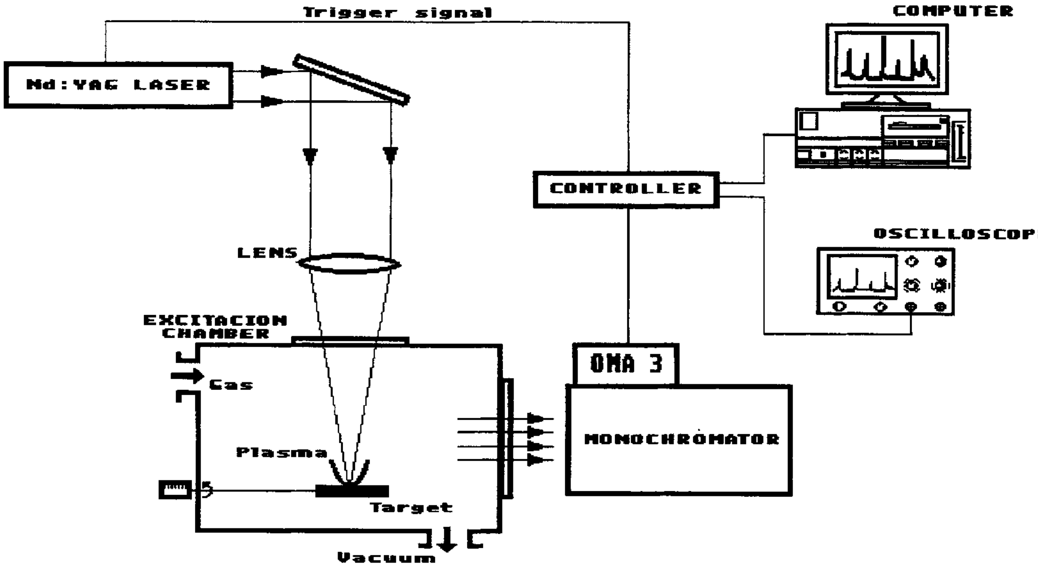

Fig. 1. Block diagram of the experimental setup.

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small continuous flow of gas to maintain the purity of the atmosphere. The buffer gas pressure value, 6 Torr, was chosen because it gives the best contrast between line emission and continuum emission.

As is known [9, 10], the temperature, the temporal evolution and the electron densities of the laser-produced laser can be controlled partially by using a suitable buffer gas. Moreover, the use of Ar and of Ca, the latter is found as an impurity in the sample used, provides various spectrum lines with well-known Stark parameters and transition probabilities, which will be useful for completing the determination of the plasma temperatures and electron densities.

The Pb sample was located inside the chamber, on top of a device capable of moving it horizontally with respect to the laser beam, focused in such a way that the plasma was formed in each measurement on the smooth surface of the target and not on the crater formed during the previous measurement.

The laser irradiance on the blank was 1.4 × 10^{10} W cm^{-2}, and the diameter of the standard crater was 0.5 mm; the spatial width of the focused laser beam was measured by recording its image with a 1024 element linear silicon diode array. The light emitted by the LPP was transmitted through a sapphire window to the input slit of a 1 m Czerny-Turner monochromator with a 2400 grooves mm^{-1} grating; the resolution of the spectroscopic system is 0.3 Å in the first order.

The spectrum was recorded by a time resolved optical multichannel analyzer (OMA3, EG&G) system, which can be used to record sections of the spectrum with a delay with respect to the laser pulse and for a selected interval of time; to obtain the best signal-to-noise ratio the measurements were made with a delay of 2.5 µs and the recording interval was 0.1 µs. The detection was performed in synchronized manner with the electronic device that regulates the laser Q-switch. In each data acquisition period a correction was made with regard to the dark signal in the absence of the laser plasma. The instrumental profile of the line was determined with a precision of 97%, the instrumental width (FWHM) being 0.11 Å for a wavelength of 3000 Å.

The same experimental system was used to study the homogeneity of the plasma but, in order to have spatial resolution, the light was focused by means of a lens on a 1 mm light guide, being able to select the point of the plasma from which the light emission is observed. The measurements were taken by scanning the plasma emission in two perpendicular directions; through the axis of the plasma with a distance from the blank in the 0.25–2.75 mm range to study the evolution of the plasma in space, and parallel to the surface of the target with a radial distance in the range of 0–1.12 mm, to determine where the different atomic species of the Pb are located in the plasma and to determine the real values of the parameters of the plasma (the intensities, the widths of the lines and the temperature of the plasma).

To calibrate the spectrum response of the experimental system in the range studied, from 2000–7000 Å, a deuterium lamp was used for the 2000–4000 Å range, and a tungsten lamp for the 3500–7000 Å range, both lamps having been previously calibrated.The calibration was also verified by means of Ar I and Ar II branching ratios, which are well-known [11–14], and which permit the comparison of the response selected in the spectrum regions centred in 2500, 3800 and 6500 Å.

The Ar II lines used have the wavelengths, in Å, 4266.53, 3968.36, 3944.27, 6684.31 and 6863.54, transition $3p^4(^3P) \rightarrow 4p\,^4D$, and 6483.08, 3844.57, 4441.8 and 4579.4, transition $3p^4(^3P) \rightarrow 4p\,^2S$. Also were using the lines with, in Å, 3948.98, 4045.96, 4181.88 and 4335.34, of the transition $4s\,[3/2]^0 \rightarrow 5p'\,[1/2]$ of the Ar I.

Various emission spectra were recorded to obtain the transition probabilities with regard to a statistical uncertainty of 3%.

To estimate the existence of possible self-absorption effects on the lines of Pb II measured, the absorption effects were calculated taking into account the temperature and electron density of the plasma.

3. Experimental results and discussion

3.1. *Emission spectrum*

The LPP emission spectrum in the visible was recorded for different delay times. As a general rule, in the first times of the evolution of the plasma, the spectrum lines appear widened, and it is hard to distinguish them from the intense bremsstrahlung continuum emission for times of approximately 0.1 µs. For a time of approximately 0.4 µs after the laser pulse, the species observed are the ionized atoms with high line intensities and widths; for longer times the line widths and intensities of the neutral and singly ionized species decrease considerably.

In our experimental conditions–with a delay time of 2.5 µs after the laser pulse, in an atmosphere of argon at 6 Torr – all the transitions of the Pb II spectrum can be observed, as can those of the Pb I as well as some of the Ar I, Ar II, and the most intense transitions of the Ca I and Ca II, because the sample used has 0.1% Calcium; Fig. 2 displays a typical emission spectrum obtained in the conditions described above. The Pb III emission lines appear at very early times when the temperature of the plasma is higher, disappearing at 0.9 µs.

Relative intensities have been measured for 30 lines of Pb II, evaluating the area under each one, for which purpose adjustments were made to the profiles observed of the lines by means of a convolution of the instrumental profile,

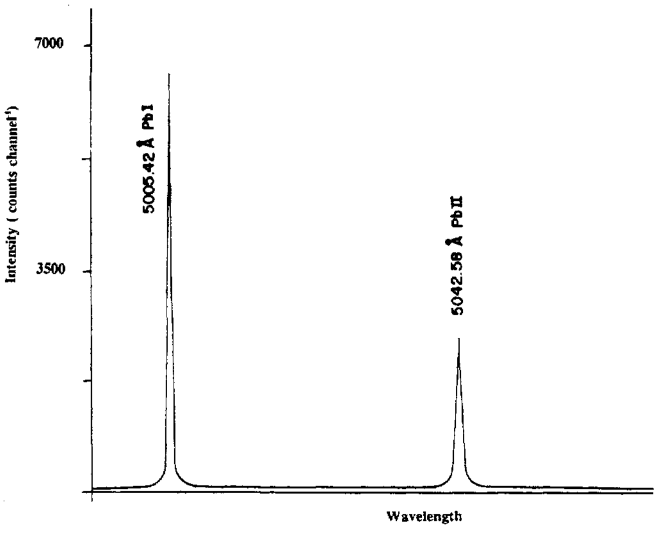

Fig. 2. Section of typical Pb II spectra at 6 Torr Ar (2.5 µs delay time).

known, with the Voigt profiles obtained from the contributions selected, Lorentz and Gaussian [15, 16]. The instrumental profile was pre-determined by observation of various narrow lines emitted by hollow cathode lamps. The setting of the profiles allows us to safely obtain the total intensities of the lines, as well as the Lorentz and Gaussian contribution in each line.

3.2. *Determination of the plasma temperature and electron densities*

When LTE conditions can be applied, the populations of the linked states follow Boltzmann's distribution, which can be used as a first approximation to determine the temperature and the electron density [17, 18]. In an optically thin plasma the relative intensities I_{ij} of the lines emitted from a given state of excitation can be used to calculate the electron temperature if the A_{ij} transition probabilities are known, by the expression:

$$I_{ij} = \frac{A_{ij} g_i}{U(T)} N \exp(-E_i/kT) \tag{1}$$

for a transition from a higher state i to a lower state j, I_{ij} is the relative intensity, E_i and g_i are the energy and statistical weight of level i, $U(T)$ is the atomic species partition function, N the total density of emitting atoms, k the Boltzmann constant and T the temperature. If we were to plot $\ln(I_{ij}/g_i A_{ij})$ *vs.* E_i, Boltzmann plot, the resulting straight line would have a slope $-1/kT$, and therefore the temperature can be obtained without having to know the total density of atoms or the atomic species partition function. The energies of the different levels are those of Moore [19].

The designations and the Pb II transition probabilities selected for determination of the excitation temperature by Boltzmann plot are shown in Table I. The relative intensities I_{ij} required for application of this method were obtained in this study and the corresponding transition probabilities are those obtained in our previous study, already mentioned above [4]. Figure 3 displays a Boltzmann plot from which a value of $(11\,500 \pm 350)$ K for $\Delta E = 3.33$ eV was obtained for the electron temperature.

As a confirmation of the LTE hypothesis, we also obtained the temperature of the plasma deduced from lines 3933.7 Å, 3706.0 Å, 3776.9 Å and 5001.5 Å of the Ca II with a value of $(11\,700 \pm 500)$ K for $\Delta E = 9.98–3.12 = 6.87$ eV; the transition probabilities for the lines of Ca II are taken from the compilation of Wiese [11]. We also obtained the temperature of the plasma deduced from the buffer gas lines; the spectrum lines selected for Ar II were 3979.4 Å, 3952.7 Å and 3968.4 Å, a value of $(11\,600 \pm 500)$ K for $\Delta E = 23.08–19.55 = 3.53$ eV being obtained. The transition probabilities for these lines of the Ar II are taken from the study by Vujnovic [14]. These values obtained for the temperature are totally compatible with the value obtained from the lines of Pb II.

Table I. *Parameters for electron temperature calculations*

Transition	λ (Å)	E_i (eV)	$A_{ij}(\times 10^6\ s^{-1})$ (Ref. [4])
$7p\,^2P^0_{1/2} \rightarrow 7s\,^2S_{1/2}$	6660.0	9.332	59.1 ± 5.9
$7p\,^2P^0_{3/2} \rightarrow 7s\,^2S_{1/2}$	5608.9	9.581	83.1 ± 8.3
$7d\,^2D_{3/2} \rightarrow 7p\,^2P^0_{1/2}$	5042.6	11.690	101.7 ± 15.2
$9s\,^2S_{1/2} \rightarrow 7p\,^2P^0_{3/2}$	4152.8	12.566	22.6 ± 3.2
$5f\,^2F^0_{5/2} \rightarrow 6d\,^2D_{3/2}$	4386.5	11.473	147.1 ± 14.7
$5f\,^2F^0_{7/2} \rightarrow 6p^2\,^4P_{5/2}$	5372.3	11.471	60.0 ± 6.1

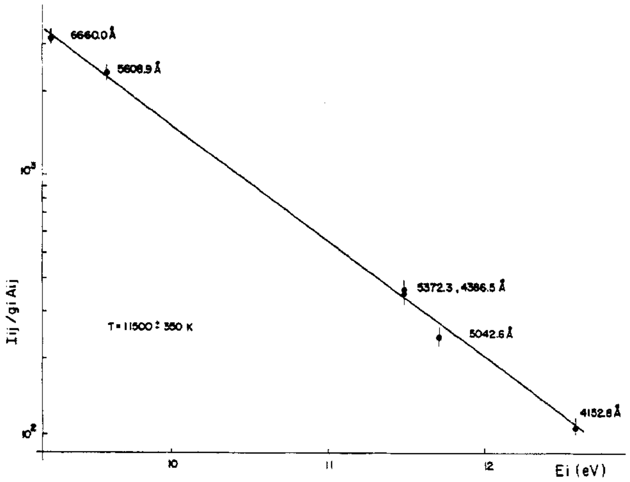

Fig. 3. Boltzmann plot for Pb II lines emitted from a laser produced plasma at 2.5 μs delay in Ar at a pressure of 6 Torr.

McWhirter's criterion, for the lower limit of the electron density, has been used to support the LTE hypothesis, and this is given by the following expression [20–21]:

$$N_e(\mathrm{cm}^{-3}) \geqslant 1.6 \times 10^{12} \sqrt{T} (\Delta E)^3 \tag{2}$$

where ΔE, in eV, is the energy difference between the upper and lower states, and T, in K, the temperature and N_e the lower limit of the electron density necessary to maintain the populations of the energy levels at 10% of the LTE by collision, in competition with the radiative processes. Using the values obtained of the lines of Pb II, the critical N_e is $0.63 \times 10^{16}\,\mathrm{cm}^{-3}$.

The electron densities, N_e, of the plasma investigated have been obtained by comparing the Stark broadenings for several transitions with those of other authors, see Table II, using the expression [20–22]:

$$\Delta\lambda = 2\omega\left(\frac{N_e}{10^{16}}\right)\left[1 + 1.75A\left(\frac{N_e}{10^{16}}\right)^{1/4}(1 - 1.2N_D^{-1/3})\right] \tag{3}$$

where $\Delta\lambda$ is the full width at half maximum (FWHM) of the transition considered, ω is the electron impact parameter, A is the ion-broadening parameter and N_D is the number of particles in the Debye sphere, which must be in excess of the lower limit $N_D = 2$ of the Debye approximation for correlation effects. For the electron densities present in this study, the ion-broadening contribution is only approximately 5% and the value of N_D estimated by $N_D = 1.38 \times 10^3 T^{3/2} N_e^{-1/2}$, [20], is much higher than the limit for application of the theory.

The electron densities obtained from the Stark broadenings may be considered reliable because the other broadening mechanisms assessed in this study barely account for 3% of the total broadening value.

The values of the electron densities from very different spectrum lines are in good agreement; Table II displays the values obtained for the 4244.9 Å, 5544.3 Å and 5608.9 Å of Pb II and for the 3933.7 Å of Ca II, with the third column giving the Stark broadenings obtained by Puric [23] for the lines of Pb II and those obtained by Goldbach [24] for

Table II. *Electron density of plasma (6 Torr Ar; delay time of 2.5 μs)*

Pb II Transition	λ (Å)	$T = 16000\,\mathrm{K}$ $N_e = 10^{17}\,\mathrm{cm}^{-3}$ $\omega_{exp} \pm 15\%$ (Ref. [23])	N_e ($10^{16}\,\mathrm{cm}^{-3}$)
$5f\,^2F^0_{7/2} \rightarrow 6d\,^2D_{5/2}$	4244.9	1.52	0.97 ± 0.20
$7d\,^2D_{5/2} \rightarrow 7p\,^2P^0_{3/2}$	5544.3	3.18	1.00 ± 0.19
$7p\,^2P^0_{3/2} \rightarrow 7s\,^2S_{1/2}$	5608.9	2.12	0.99 ± 0.16

Ca II Transition	λ (Å)	$T = 11700 \pm 600\,\mathrm{K}$ $N_e = 10^{17}\,\mathrm{cm}^{-3}$ $\omega_{exp} \pm 30\%$ (Ref. [24])	N_e ($10^{16}\,\mathrm{cm}^{-3}$)
$4p\,^2P^0_{1/2} \rightarrow 4s\,^2S_{1/2}$	3933.7	0.10	1.16 ± 0.36

the line of Ca II. The energy interval, ΔE, for the temperatures and electron densities ranges from $\Delta E = 3.33\,\mathrm{eV}$ for Pb II, $\Delta E = 3.53\,\mathrm{eV}$ for Ar II and $\Delta E = 6.87\,\mathrm{eV}$ for Ca II.

With the aforementioned values of N_e and T we can calculate the absorption coefficient for the lines studied, using equation for the absorption coefficient, expressed in m^{-1}:

$$k_\omega = \frac{\pi e^2}{2\varepsilon_0 mc} f_{ik} N_i [1 - (N_k g_i / N_i g_k)] g(\omega). \tag{4}$$

f_{ik} is the oscillator strength (absorption) and $g(\omega)$ the profile of the line. In the maximum, $\omega = 0$, and for a Lorenz profile, $g(0) = 2/\pi\Gamma$, where Γ is the FWHM of the line; a line may be considered optically thin if $k_\omega L \leqslant 0.05$, [25], checking that the value of the optical depth $k_\omega L$ in this work is not in excess of 0.02; for example 0.019 in 6660.0 Å, 0.015 in the 5608.9 Å or 0.006 in the 5042.6 Å. These values are less than the lower limit, confirming which the optical thin hypothesis used to be deemed valid; the lines studied in this study are not resonant.

All of this confirms the LTE hypothesis for the levels of Pb II considered in this study, which are strongly coupled by radiation and separated by energy intervals of less than 4 eV.

The values given for N_e and T correspond to the centre of the plasma; to determine the change of these parameters in different regions of the plasma, we have obtained their values at different points using various lines of Pb II, and the result being that there is homogeneity for N_e and T; deviations from the average are less than 15% for N_e and 5% for T in a region measuring approximately 2 mm in size corresponding to 95% of the emission of light; similar results have been obtained in other LPP experiments [7, 9, 10].

3.3. *Pb II transition probabilities obtained from Boltzmann plot*

The transition probabilities obtained from Boltzmann plot for the 30 lines of Pb II with wavelengths in the range 2700–6800 Å are displayed in column three of Table III,

Table III. *Pb II transition probabilities obtained from Boltzmann plot*

		Absolute transition probabilities ($\times 10^6\,\mathrm{s}^{-1}$)				
		Experimental			Theory	
Transition levels	λ (Å)	This work	Ref. [4]	Ref. [1]	Ref. [4]	Ref. [26]
$8s\,^2S_{1/2} \rightarrow 7p\,^2P^0_{1/2}$	6791.2	44.7 ± 4.5			52.2	38.0
$9s\,^2S_{1/2} \rightarrow 7p\,^2P^0_{1/2}$	3718.3	12.0 ± 2.0	17.5 ± 2.8		16.3	14.1
$7p\,^2P^0_{3/2}$	4152.8	22.3 ± 2.8	22.6 ± 3.6	24 ± 10	23.4	29.9
$10s\,^2S_{1/2} \rightarrow 7p\,^2P^0_{1/2}$	2986.9	7.6 ± 0.8	8.4 ± 1.2		7.3	8.1
$7p\,^2P^0_{3/2}$	3260.9	12.9 ± 1.9	10.4 ± 1.5		11.3	12.4
$7p\,^2P^0_{1/2} \rightarrow 7s\,^2S_{1/2}$	6660.0	57.2 ± 5.4	59.1 ± 5.9	62 ± 21	56.3	73.8
$6p^2\,^4P_{1/2}$	6041.4	0.54 ± 0.07	0.62 ± 0.06			
$7p\,^2P^0_{3/2} \rightarrow 7s\,^2S_{1/2}$	5608.9	84.8 ± 8.5	83.1 ± 8.3	124 ± 43	94.4	124.9
$6p^2\,^4P_{1/2}$	5163.3	1.1 ± 0.2	1.6 ± 0.2			
$8p\,^2P^0_{1/2} \rightarrow 7s\,S_{1/2}$	2805.9	5.6 ± 0.6			4.4	1.8
$6p^2\,^4P_{1/2}$	3945.7	0.51 ± 0.06			0.44	
$8p\,^2P^0_{3/2} \rightarrow 7s\,^2S_{1/2}$	2717.4	7.3 ± 0.8			4.9	14.0
$6p^2\,^4P_{1/2}$	3665.5	0.56 ± 0.06			0.46	
$7d\,^2D_{3/2} \rightarrow 7p\,^2P^0_{1/2}$	5042.6	90.0 ± 8.9	101.7 ± 15.2	77 ± 25	106.7	
$7p\,^2P^0_{3/2}$	5876.6	13.8 ± 1.0	16.8 ± 2.5		13.5	
$7d\,^2D_{5/2} \rightarrow 7p\,^2P^0_{3/2}$	5544.3	102.8 ± 10.6		104 ± 31	96.6	
$8d\,^2D_{3/2} \rightarrow 7p\,^2P^0_{1/2}$	3455.1	42.1 ± 6.1	43.7 ± 7.0		42.9	
$7p\,^2P^0_{3/2}$	3827.2	6.3 ± 0.7	5.8 ± 0.9		6.3	
$8d\,^2D_{5/2} \rightarrow 7p\,^2P^0_{3/2}$	3713.9	41.9 ± 6.2			42.2	
$5f\,^2F^0_{5/2} \rightarrow 6d\,^2D_{3/2}$	4386.5	155.7 ± 15.6	147.1 ± 14.7	44 ± 15	167.7	
$6d\,^2D_{5/2}$	4242.1	9.8 ± 1.0	9.3 ± 0.9		12.6	
$6p^2\,^4P_{3/2}$	3785.9	10.1 ± 1.0	9.6 ± 1.4	3.3 ± 1.4		
$6p^2\,^4P_{5/2}$	5367.6	3.1 ± 0.3	3.0 ± 0.3			
$5f\,^2F^0_{7/2} \rightarrow 6d\,^2D_{5/2}$	4244.1	115.0 ± 11.6	112.1 ± 16.8	29 ± 10	133.7	
$6p^2\,^4P_{5/2}$	5372.3	62.1 ± 6.2	60.0 ± 6.1	128 ± 38		
$6f\,^2F^0_{5/2} \rightarrow 6d\,^2D_{3/2}$	3016.4	40.9 ± 5.0	41.1 ± 6.2		41.8	
$6d\,^2D_{5/2}$	2947.4	3.3 ± 0.4	3.5 ± 0.5		3.1	
$6p^2\,^4P_{3/2}$	2719.8	1.7 ± 0.2				
$6f\,F^0_{7/2} \rightarrow 6d\,^2D_{5/2}$	2948.5	44.8 ± 4.9			47.0	
$6p\,^4P_{5/2}$	3451.7	5.5 ± 0.6				

while columns one and two give the transitions and corresponding wavelengths respectively. The remaining columns give the theoretical [4, 26] and experimental [1, 4] transition probability values to be found in the bibliography; column four gives those obtained in the study [4] previous to this one, and already mentioned above, and a good match is observed between the values in obtained in both.

4. Conclusions

In this study we have obtained the transition probability values in 30 lines of the spectrum of emission of Pb II, in 9 of which they are the first experimental data that are obtained. It is seen that, under certain experimental conditions, and as far as temperature and electron density are concerned, Laser Produced Plasmas (LPP) is a homogenous and stable enough source to allow us to measure the atomic transitions probabilities of ionized species, even though this calls for an experimental system with a good time resolution.

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