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Stark widths of several Pb III spectral lines in a laser-induced lead plasma

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ABSTRACT

Context. Data on Stark widths of spectral lines are relevant not only for atomic structure research, but also for astrophysics and analytical techniques of plasma diagnosis.
Aims. Stark widths of ten doubly ionized lead spectral lines that belong to 7s–7p, 6d–7p and $6p^2$–7p transitions have been measured.
Methods. We observe the emission lines from an optically thin laser induced plasma from a lead target in an argon atmosphere. The studied plasma has a temperature of about 25 200 K and an electron density of about 10^{17} cm^{-3}. Local thermodynamic equilibrium conditions and plasma homogeneity have been checked. Special attention was paid to the possible selfabsorption of the different transitions.
Results. We report ten new experimental values for Stark widths (*FWHM*) of Pb III lines.

Key words. stellar dynamics – plasmas – line: profiles

1. Introduction

Data on Stark widths of spectral lines are relevant not only for atomic structure research, but also for astrophysics and analytical techniques of plasma diagnosis.

The combination of the Hubble Space Telescope (HST) and the Goddard High Resolution Spectrograph (GHRS) has allowed the discovery of a number of elements heavier than zinc in the interstellar gas. With $Z = 82$, lead has the highest cosmic abundance among the elements heavier than barium.

The analysis of spectral lines of doubly ionized lead, Pb III, could allow us (with the new generation telescopes such as, NASAs Next Generation Space Telescope (NGST)) to obtain information about the physical conditions of the hot stars. Knowledge of the Stark widths of the spectral lines enables modeling of various physical processes in the hot star plasmas where the Stark broadening is the principal pressure broadening mechanism.

In this paper, Stark widths of 10 spectral lines corresponding to the 7s–7p, 6d–7p and $6p^2$–7p transitions of Pb III have been measured. These values are the first experimental data in the literature.

The application of laser ablation for analysis of a solid sample is one of the most important applications of laser induced plasma (LIP). 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. From the experimental point of view, LIPs have proved to be a valuable source of spectroscopic data on neutral and ionized species (Irons 1973; Zhao et al. 1992; Alonso-Medina 1997; Colón et al. 1999; Alonso-Medina & Colón 2000; Alonso-Medina et al. 2001; Alonso-Medina et al. 2003; Harilal et al. 2005; Colón & Alonso-Medina 2006; ...).

In the present experiment, a laser induced plasma on a lead surface was used as a spectral source. Experimental working conditions of stability and homogeneity of electron density and temperature in the plasma were determined by means of a study of the temporal evolution in different environmental conditions and target composition.

The present measurements were carried out with a lead target of 99.99% purity placed in an argon atmosphere at 12 Torr and 0.4 μs after each laser light pulse. We will describe in Sect. 2 the experimental system used for LIP and the emission spectrum studied. The results obtained are given in Sect. 3, and the conclusions in Sect. 4.

2. Experimental setup and emission spectrum

A plasma was generated by focusing a laser beam on a sample lead surface. The experimental system has been described in previous papers (Alonso-Medina 1997; Colón et al. 1999; Alonso-Medina et al. 2001; Colón & Alonso-Medina 2006).

A chamber was used to generate the plasma in a gas atmosphere. A vacuum of 10^{-5} Torr was attained inside the chamber by means of a turbomolecular pump, and it was filled with argon and maintained at a constant pressure of 12 Torr throughout the measurement, using a small continuous flow of gas to maintain the purity of the atmosphere. In this way the temperature, the electron density and the temporal evolution of LIP could be controlled. In Fig. 1 we present sections of typical spectra.

Samples were 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 beam used to generate the plasma was produced by a Q-switched Nd:YAG laser (Quantel YG585), that generates 290 mJ pulses of 7 ns duration at a frequency of 20 Hz and 10 640 Å wavelength. Its beam was focused on the sample by a lens of focal distance of 12 cm. The laser irradiance on the blank was 1.4×10^{10} W cm^{-2},

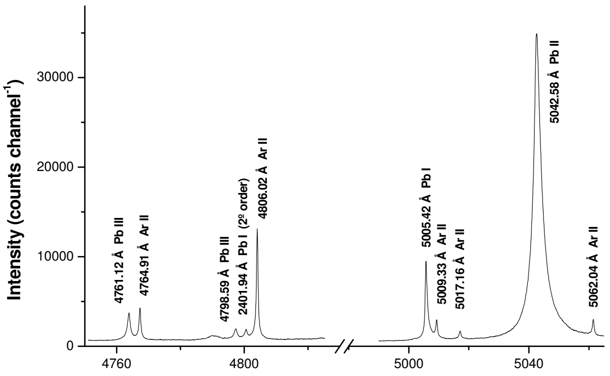

Fig. 1. Emission lines of Pb I, Pb II, Pb III and Ar II at 12 Torr in argon at 0.4 μs delay time.

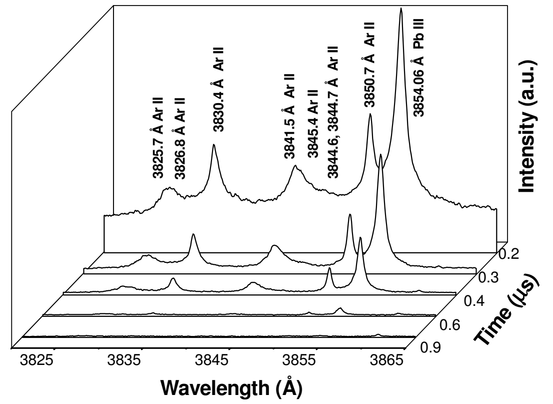

Fig. 2. Time resolved emission spectra from laser produced lead plasma in 12 Torr of argon.

producing craters with standard diameters of 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 LIP was transmitted, through a sapphire window, to the input slit of a 1 m Czerny Turner spectrometer provided with a 2400 grooves mm^{-1} holographic grating. The resolution of the spectroscopic system was 0.3 Å in first order.

The spectral response of the system was obtained in the 1900 to 7000 Å wavelength range by means of previously calibrated lamp. A deuterium lamp was used for the 1900 to 4000 Å range and a tungsten lamp for the 3500 to 7000 Å range. The estimated error of this measurement was about 3%. The calibration was also verified by means of Ar I and Ar II branching ratios to permit the comparison of the response selected in the spectral regions centred at 2500, 3800 and 6500 Å (Alonso-Medina 1997). In order to obtain a statistical uncertainty of 3% various emission spectra were obtained and analyzed.

Spectra were recorded by a time-resolved optical multichannel analyser (OMA III EG&G) that allows the recording of spectra at a preset delay from the laser pulse and with a selected time length. Spectra were obtained at 0.1 to 9.0 μs delay from the laser pulse and light was collected for 0.1 μs in synchronism with the electronic trigger of 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 optical device contribution to the total broadening of the spectral line (instrumental profile) was determined by observation of various narrow lines emitted by hollow cathode lamps. The spectra were stored in a computer for further analysis, which was made by fitting the observed line shapes to numerically generated Voigt profiles.

The same experimental system was used to study the homogeneity of the plasma but, in order to have sufficient spatial resolution, the light was focused by a lens on a 1 mm light guide being able to select the point of the plasma from which the light emission was observed. The measurements were taken by scanning the plasma emission in two perpendicular directions, as can be seen in Fig. 1 of Alonso-Medina et al. (2001), to determine where the different atomic species of lead are located in the plasma and to determine the real values of the parameters of the plasma. Local profiles were obtained after Abel inversion of the integrated intensity (Lochte-Holtgreven 1968). The fitting of the observed profiles provides the total intensity very accurately, as well as the broadening of the spectral lines.

The LIP emission spectrum was recorded in the argon atmosphere for different delay times. As a general rule, early in 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 widths and intensities of the neutral and singly ionized species decrease considerably.

In Fig. 2 time resolved emission spectra at 0.2 to 0.9 μs from laser produced plasma are presented. In the argon atmosphere, at short times the spectral emission lines of Ar II and Pb III appear. But the Pb III emission lines appear at very early times when the temperature of the plasma is higher, disappearing at 0.9 μs.

The lead sample has a purity of 99.99%. In our experimental conditions with a delay time of 0.4 μs after the laser pulse in an atmosphere of argon at 12 Torr all the transitions of the Pb III spectrum can be observed, as can those of Pb I, Pb II and Ar II. Relative intensities have been measured evaluating the area under each line. For this purpose adjustments were made to the profiles observed using a convolution of the known instrumental profile, with the Voigt profile, obtained from the Lorentzian and Gaussian contributions (Bruggemann & Bollig 1992).

3. Experimental results

As is well known, in 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

$$I_{ij} = \frac{A_{ij} g_i}{U(T)} N \exp\left(\frac{-E_i}{kT}\right), \qquad (1)$$

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, the 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 (1958, 1971).

The plasma temperature has been determined by means of a Boltzmann plot for some lines of Pb II, whose transitions probabilities are known (Alonso-Medina 1997), and was found to be 25 400 ± 1500 K; for some lines of Pb III, the transition probabilities (Colón et al. 1999) were used to obtain the temperature of

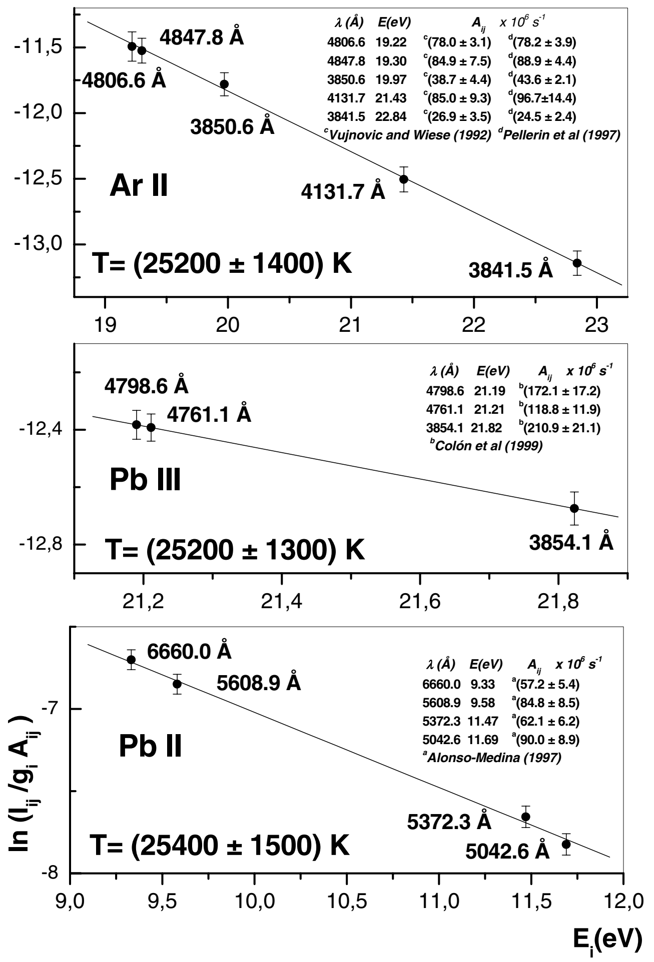

Fig. 3. Boltzmann plots for Pb II, Pb III and Ar II lines from laser produced plasma at 0.4 μs at a pressure of 12 Torr in argon.

25 200 ± 1300 K and with some lines of Ar II (Vujnović & Wiese 1992; Pellerin et al. 1997), the plasma temperature was found to be 25 200 ± 1400 K. These Boltzmann plots are presented in Fig. 3. These values are totally compatibles.

The electron density, N_e, of the plasma has been obtained by comparing the Stark broadening for several transitions with those of other authors, using the expression by Griem (1964) and Milosavljević & Poparić (1999)

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

where ω is the full width al half maximum (FWHM) of the transition considered and obtained at the density N_e expressed in cm^{-3}, ω_p is the Stark broadening parameter (full half-width for the density 10^{16} cm^{-3}), 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. In our measurements we have assumed that A is negligible (Konjević 2001).

The value of the electron density was obtained using in the last equation the experimental value of the Stark broadening parameter, ω_p, for the 4244.9 Å Pb II transition. This value of ω_p = 1.50 Å was obtained for a temperature of 24 000 K and $N_e = 1.0 \times 10^{17}$ cm^{-3} (Salakhov et al. 1984). The Stark broadening of this transition was obtained as the Lorentzian part, ω = 1.5 Å, of its Voigt profile. The electron density was $(1.01 \pm 0.19) \times 10^{17}$ cm^{-3}.

Table 1. Electron density of plasma (12 Torr of argon, delay time 0.4 μs).

Atomic specie	λ (Å)	T (K)	ω_p(Å) $N_e = 10^{16}$ cm^{-3}	N_e(10^{17} cm^{-3}) 25 400 K
Pb II				
	4244.9	20 000	0.13^a ± 15%	1.01 ± 0.19
		24 000	0.15^b ± 15%	
	5544.3	20 000	0.21^a ± 15%	1.10 ± 0.11
	5608.9	20 000	0.17^a ± 15%	1.00 ± 0.18
Ar II				
	4806.0	31 000	0.04^c ± 30%	1.15 ± 0.36

[a] Purić et al. (1985); [b] Salakhov et al. (1984); [c] Roberts (1968).

The values of the electron densities from very different spectrum lines are in good agreement. The Stark broadening parameters for these lines were obtained by extrapolation in Table 1 which displays the values obtained with the 5608.9, 5544.3 and 4244.9 Å lines of Pb II and also 4806.0 Å of Ar II. The fourth column displays the Stark broadening measured by Purić et al. (1985), Salakhov et al. (1984) and Roberts (1968). The fifth column displays the electron densities obtained using the values of the previous column, normalized to the temperature, 25 400 K, of our plasma.

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

The obtained value of 10^{17} cm^{-3} is sufficient to assume LTE for the population of the studied levels according to the criterion of McWhirter (1963)

$$N_e \left(\mathrm{cm}^{-3}\right) \geq 1.6 \times 10^{12} \sqrt{T} (\Delta E)^3, \qquad (3)$$

were ΔE, in eV, is the energy difference between both levels and T, in K, the plasma temperature, and N_e the lower limit of the electron density necessary to maintain the populations of the energy level at 10% of the LTE by collision, in competition with the radiative processes.

Using the values obtained for the lines of Pb II the critical N_e is 3.3×10^{15} cm^{-3}, using the values obtained for the lines of Pb III the critical N_e is 6.4×10^{13} cm^{-3}, and using the values obtained of the line of Ar II the critical value is 1.2×10^{16} cm^{-3}.

As a further confirmation of the LTE hypothesis, in our experimental conditions for a delay time of 0.4 μs after the laser pulse, in an atmosphere of Ar at 12 Torr by applying the Saha equation to the Pb I line at 5005.5 Å and the 5042.6 Å and 5602.9 Å lines of Pb II, using the transition probabilities given by Alonso-Medina (1997, 2001), we found a value of 25 300 ± 1500 K.

With the aforementioned values of N_e and T we can calculate the absorption coefficients (Thorne 1988). In our experimental conditions the value of the optical absorption coefficients are negligible.

In this work the spectroscopic analysis of the laser produced plasma emission provides experimental Stark broadening parameters for 10 new lines of Pb III. Table 2 shows the results of our experimental data of full width half maximum (*FWHM*) parameters normalized to an electron density of 10^{17} cm^{-3}. The corresponding errors include uncertainties in the instrumental

Table 2. Experimental Stark Width FWHM, ω(Å), of emission lines of Pb III normalized at $N_e = 10^{17}$ cm^{-3} (12 Torr of argon, delay time 0.4 μs and 25 200 K).

Transition array	Multiplet	λ(Å)	ω(Å)
7s–7p	$^3S_1 - ^3P_0$	4798.59	1.0 ± 0.2
	$^3S_1 - ^3P_1$	4761.12	1.0 ± 0.2
	$^3S_1 - ^3P_2$	3854.08	0.75 ± 0.16
	$^1S_0 - ^3P_1$	5779.41	2.9 ± 0.7
6d–7p	$^1D_2 - ^3P_1$	5207.10	2.2 ± 0.6
	$^1D_2 - ^3P_2$	4141.59	2.7 ± 0.6
	$^3D_1 - ^3P_2$	5380.12	2.7 ± 0.7
	$^3D_2 - ^3P_2$	5523.97	2.4 ± 0.6
	$^3D_3 - ^3P_2$	5857.96	1.1 ± 0.3
6p^2–7p	$^3D_1$ - $^3P_2$	4855.06	1.4 ± 0.4

profile and statistical errors after an average of several spectra with a total of about 100 laser shots. The possible error due to the experimental uncertainty in the density of electrons in this study is also included. The first three columns denote the corresponding transition array, the multiplet and the wavelengths (in Å) for each studied transition. The fourth column compiles our experimental Stark width parameters.

4. Conclusions

1. we report ten new experimental values for Stark width (FWHM) of Pb III lines;
2. optical emission spectroscopy is used to characterize the laser produced plasma from a lead target in an argon atmosphere. Time and space resolved measurements of electron density and temperature have been carried out. At 0.4 μs delay time from the laser pulse there are adequate experimental conditions to measure the atomic parameters of Pb III;
3. the homogeneity of the lead plasma has been studied and a temperature of 25 200 K and an electron density close to 10^{17} cm^{-3} has been obtained. No self-absorption effects were detected.

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