Lifetime measurements and calculations in La I

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Abstract. Radiative lifetime measurements have been performed, with a time-resolved laser-induced fluorescence technique, for 20 odd-parity levels of La I belonging to the configurations $5d^26p$, 5d6s6p and 4f5d6s. The new results are compared with the few experimental data available in the literature and with theoretical calculations including configuration interaction effects. The agreement theory-experiment is generally satisfying but discrepancies are observed for some levels emphasizing the difficulty to get a reliable theoretical model in such a heavy and complex neutral element.

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1 Introduction

Radiative parameters of spectral lines (i.e. transition probabilities, radiative lifetimes and branching fractions) are needed in astrophysics, particularly for the lanthanide group, in order to analyse quantitatively the high resolution stellar spectra now available and to refine our knowledge of the chemical composition of some celestial objects. In particular, the study of chemically peculiar stars, which are characterized by strong overabundances of the rareearth elements when compared to the solar system values, needs a large amount of new accurate data for the atoms in their first three ionisation stages which, according to the Saha equation, are expected to occur in ionisation equilibrium in such stars. Infrared and visible observations of chemically peculiar stars (showing mostly lines of neutrals or of neutrals and ions, respectively) are complementary because spectral lines in these wavelength ranges allow to probe different depths in stellar atmospheres and to shed some light upon the relations between inhomogeneities and abundance anomalies.

La I has two stable isotopes, i.e. 138 La (0.09%) and 139 La (99.91%) with nuclear spins 3/2 and 7/2, respectively. The ground-state level of neutral lanthanum is $5d6s^2 \ ^2D_{3/2}$. According to the NIST compilations ([1]; see also the web site http://www.physics.nist.gov/cgi-bin/AtData/levels_form), our knowledge of both the energy levels and radiative parameters of La I is still very fragmentary and calls for many additional efforts. The energy levels of La I, as compiled by the NIST, based on early analyses of this spectrum, are belonging to the configurations $5d6s^2$, $5d^26s$, $5d^3$, 4f6s6p, $5d^27s$,

5d6s7s, and $5d^26d$ for the even parity and to 5d6s6p, $4f6s^2$, $6s^26p$, $5d^26p$, 4f5d6s, $5d^27p$, $4f5d^2$, 5d6s7p and $6s^28p$ for the odd parity. Above the ionisation threshold, some members of the Rydberg series $6s^2np$ (n = 9-23)have been determined. It must be stressed that the designation of some levels is still uncertain and that most of them show dramatic configuration interaction mixings. In addition, many levels in both parities (particularly above $33\,000$ cm⁻¹) are not yet assigned.

Measurements of transition probabilities in La I were first published by Corliss and Bozman [2] about forty years ago. These measurements are known to be affected by large systematic errors, particularly for the weaker transitions. Lifetime measurements, for a few levels belonging to the 5d6s6p configuration, are due to Hese and coworkers [3-5] who used level-crossing and doubleresonance techniques, to Bulos et al. [6] who measured the z ²F_{5/2}, z ²D_{3/2}, z ⁴G_{5/2} and y ²D_{3/2} levels by the laserinduced fluorescence method and to Penkin et al. [7] who considered the pulse-electron delayed coincidence technique. Discussions concerning the lifetimes or transition probabilities for the resonance transitions in La I are due to Blagoev and Komarovskii [8] and to Doidge [9], respectively. Oscillator strengths, deduced from the solar spectrum, have been published by Thevenin [10] in an attempt to supply some missing data and a compilation of astrophysical interest, including La I, is due to Morton [11].

In view of the scarcity of the radiative data available and of the large uncertainties affecting some of them, the purpose of the present work is to obtain new reliable lifetimes for levels of La I emitting lines of astrophysical interest. The obtention of such data is the first step for deducing accurate transition probabilities which rely,

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Configuration	Term	J	$E ({\rm cm}^{-1})$	Exci	tation	Laser*	Observed	
				origin	$\lambda(nm)_{vac.}$	mode	$\lambda(\mathrm{nm})_{vac}$	
$5d^{2}(^{3}\mathrm{F})6p$	$^{2}\mathrm{D}^{\circ}$	3/2	18172.35	0.0	550.286	$2\omega + 3S$	550	
$5d^2(^3\mathrm{F})6p$	$^{2}\mathrm{D}^{\circ}$	5/2	19379.40	1053.164	545.665	$2\omega+3S$	545, 516	
$5d^2(^3\mathrm{F})6p$	$^{4}\mathrm{D}^{\circ}$	1/2	22246.64	0.0	449.506	$2\omega+2S$	511	
$5d^2(^3\mathrm{F})6p$	$^{4}\mathrm{D}^{\circ}$	3/2	22439.36	0.0	445.645	$2\omega+2S$	515	
$5d^2(^3\mathrm{F})6p$	$^{4}\mathrm{D}^{\circ}$	5/2	22804.25	3494.526	517.874	$2\omega + 3S$	518	
$5d^2(^3\mathrm{F})6p$	$^{4}\mathrm{D}^{\circ}$	7/2	23303.26	1053.164	449.436	$2\omega+2S$	521	
$5d6s(^{3}\mathrm{D})6p$		7/2	23221.10	4121.572	523.573	$2\omega + 3S$	524	
$5d6s(^{3}\mathrm{D})6p$		5/2	23874.95	0.0	418.849	$2\omega+2S$	419	
$4f5d(^{3}\mathrm{F}^{\circ})6s$	${}^{4}\mathrm{F}^{\circ}$	5/2	24507.87	1053.164	426.353	$2\omega+2S$	408, 426	
$4f5d(^{3}\mathrm{F}^{\circ})6s$	${}^{4}\mathrm{F}^{\circ}$	7/2	25380.27	3010.002	447.021	$2\omega+2S$	457	
$4f5d(^{3}\mathrm{F}^{\circ})6s$	${}^{4}\mathrm{F}^{\circ}$	9/2	25997.17	3494.526	$444.392 2\omega + 2S$		457	
$4f5d(^{1}\mathrm{G}^{\circ})6s$		7/2	24409.68	1053.164	428.146	$2\omega+2S$	428	
$5d^2(^3\mathrm{P})6p$	$^{4}\mathrm{D}^{\circ}$	7/2	25083.36	1053.164	416.143	$2\omega+2S$	416	
$5d^2(^{3}\mathrm{P})6p$	$^{2}\mathrm{P}^{\circ}$	3/2	27225.26	0.0	367.305	$2\omega + S$	367, 495	
$5d^2(^1D)6p$		5/2	27393.04	0.0	365.056	$2\omega + S$	365	
$4f5d(^{3}\mathrm{G}^{\circ})6s$	${}^{4}\mathrm{G}^{\circ}$	7/2	27455.31	3494.526	$417.348 2\omega + 2S$		409, 417	
$4f5d(^{3}\mathrm{F}^{\circ})6s$	$^{2}\mathrm{F}^{\circ}$	5/2	27669.37	0.0	361.410	$2\omega + S$	361	
$4f5d(^{3}\mathrm{F}^{\circ})6s$	$^{2}\mathrm{F}^{\circ}$	7/2	28543.08	1053.164	363.769	$2\omega + S$	364, 488	
$5d6s(^{1}\mathrm{D})6p$		7/2	28039.45	1053.164	370.558	$2\omega + S$	371	
$5d6s(^{3}\mathrm{D})6p$		5/2	28506.41	1053.164	364.255	$2\omega + S$	364	

Table 1. La I levels measured (energies in cm^{-1}) and excitation schemes.

* 2ω means frequency doubling, S, 2S and 3S represent the first-, the second- and the third-order Stokes components. The energy level values are taken from Martin et al. [1].

in many cases, upon the combination of lifetime measurements and theoretical or experimental branching fraction determinations. Using a selective laser excitation, new lifetimes have been measured for 20 odd-parity levels of La I. For most of them, there were no data previously available. This work is an extension of similar measurements carried out recently in La II [12] and La III [13].

2 Lifetime measurements

The radiative lifetimes of 20 odd-parity levels, belonging to the $5d^26p$, 5d6s6p and 4f5d6s configurations, were measured using the method of time-resolved laser-induced fluorescence (for a detailed description of the experimental set-up, see e.g. Xu et al. [14]). The level values are taken from the NIST atomic spectra database [1] (http://www.physics.nist.gov/cgi-bin/ AtData/levels_form). The relevant information on the levels, on the excitation schemes and on the corresponding observed wavelengths is collected in Table 1.

Two Q-switched Nd:YAG lasers were used in our experiment. One of the Nd:YAG lasers (Continuum Surelite), characterised by a 10 ns pulse duration and a 2-10 mJ pulse energy at 532 nm, was used as an ablation laser to hit a pure lanthanum foil rotating in a $10^{-6}-10^{-5}$ mbar vacuum chamber and to produce a

plasma containing neutral as well as ionised lanthanum atoms. The second Nd:YAG laser (Continuum NY-82) is an injection seeded laser and provides 8-ns pulse duration and 400-mJ pulse energy at 532 nm. The laser beam was compressed to about 1 ns by a stimulated Brillouin scattering (SBS) compressor [15]. This beam of shorter pulses pumped a dye laser (Continuum Nd-60) operating with the DCM dye. To obtain the desired radiation, we used a KDP crystal and Raman shifting techniques in a H_2 cell to extend the tunable range of the laser radiation. The excitation pulse was sent horizontally into the vacuum chamber to interact with the plume about 10 mm above the foil. The ablation and the excitation pulses were synchronised by external triggering from a digital delay generator (Stanford Research Systems Model 535). Fluorescence emitted was collected by a fused-silica lens, filtered by a 1/8-m monochromator and finally detected by a Hamamatsu 1564U photomultiplier tube with a rise time of 0.2 ns. Transient signals were acquired by a Tektronix TDS 384B oscilloscope and transferred to a computer for the lifetime evaluations.

For each level investigated, the fluorescence signals in the different decay channels from excited upper levels to possible lower levels were checked in order to make sure that the La I transitions of interest were indeed studied. The fluorescence emitted by the different lanthanum ions can be identified by adjusting the delay time between the



Fig. 1. Recording of the timeresolved fluorescence signals for the $25\,083.36 \text{ cm}^{-1}$ level (asterisks) and for the $28\,506.41 \text{ cm}^{-1}$ level (circles) in La I. The excitation laser pulse (triangles) is also shown. The solid lines show an exponential fitting curve for the $25\,083.36 \text{ cm}^{-1}$ level that gives a lifetime value of 21.1(0.9) ns and a convolution fitting curve for the $28\,506.41 \text{ cm}^{-1}$ level that provides a lifetime of 7.9(0.3) ns.

excitation and the ablation pulses [16]. The strongest transition in the fluorescence emission was usually recorded and used for the evaluation of the radiative lifetime. In the course of the experiment, a series of measurements were performed, under different experimental conditions, to eliminate or minimise the possibility of systematic errors. They concerned the delay time, the intensity of the excitation and of the ablation lasers. The plasma density and atomic/ionic speeds at the observed spot can be adjusted by changing the ablation pulse energy, the size of the focused ablation pulse on the foil, the distance above the target surface, and the delay time between the ablation and excitation pulses. To check for possible collisional quenching and radiation trapping effects, the delay time between the ablation and the excitation pulses was changed from 3 to 10 μ s, and the detected fluorescence intensity was then decreased by a factor of 10, but still reasonably good signals were obtained for evaluating the lifetime values, which were found to be well unaffected. This indicated that radiation trapping and collisional quenching effects were in fact negligible. For a more detailed discussion the reader is referred to Xu et al. [17]. A pair of Helmholtz coils, providing a static magnetic field of about 100 Gauss, was added to or removed from the interaction zone to observe possible quantum beats due to the Zeeman effect. No such effect was observed. The position and width of the entrance slit of the monochromator and the delay time were adjusted during the experiment in order to identify and eliminate possible influence of flight-out-of-view effects. To ensure a linear response of the detection system, the fluorescence signals were detected with different neutral density filters inserted in the exciting laser light path.

The temporal shape of the excitation pulse had to be considered for the 28506.41 cm^{-1} level, which has a

shorter lifetime ($\tau = 7.9 \pm 0.3$ ns). While the ablation laser was turned off, a metal rod was inserted into the interaction zone of the excitation laser and the plasma, and the scattered light due to the excitation pulse was collected. The lifetime was evaluated by fitting the experimental fluorescence decay curve to a convolution of the detected excitation pulse with a pure exponential function. For the other levels, a least-squares exponential method was adopted to fit the fluorescence curves. Typical experimental curves for the levels at 25 083.36 and 28 506.41 cm⁻¹, respectively, showing the exponential and the deconvolution fitting procedures, are illustrated in Figure 1.

Every decay curve was obtained by averaging fluorescence photons emitted during 1000 pulses, in order to obtain a sufficiently smooth curve for evaluating the lifetimes. For each level measured, about 9 fluorescence decay curves were recorded under different experimental conditions. The averaged lifetime value was adopted as the final result. All the experimental lifetimes measured are reported in Table 2, together with the previous results and theoretical calculations.

3 HFR calculations

The unique properties of the lanthanides in general, and also of lanthanum in particular, result from the small radius of the 4f orbital which is smaller than that of the 5s electron. The collapse of the 4f orbital from a smallquantum-defect, large-radius hydrogenic orbital to a largequantum-defect, small-radius non-hydrogenic orbital does occur at lanthanum (Z = 57) (see, e.g. the figures in the review paper by Biémont and Quinet [18]) with the consequence that accurate calculations of wavefunctions in this atom are extremely complex. Part of the complexity

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Table 2. Observed and calculated lifetime values for La I and comparison with previous results.

Config.	Term	J	$E \ (\mathrm{cm}^{-1})$	Lifetime (ns)		ne (ns)		Landé factor	
				This work	HFR		Previous results		
					А	В		$\operatorname{Exp.}^{d}$	Theor.
$5d^{2}(^{3}\mathrm{F})6p$	$^{2}\mathrm{D}^{\circ}$	3/2	18172.35	17.7(1.4)	13.79	14.13	$14.2(0.9)^a, 18(3)^b, 16(1)^c$	0.799	0.835
$5d^{2}(^{3}F)6p$	$^{2}\mathrm{D}^{\circ}$	5/2	19379.40	17.2(1.0)	13.21	13.59	$15.2(0.9)^a, 16(1.5)^c$	1.186	1.192
$5d^2(^3\mathrm{F})6p$	$^{4}\mathrm{D}^{\circ}$	1/2	22246.64	10.1(0.9)	9.11	9.29		0.04	0.025
$5d^{2}(^{3}F)6p$	$^{4}\mathrm{D}^{\circ}$	3/2	22439.36	10.2(0.5)	9.39	9.40		1.192	1.196
$5d^{2}(^{3}F)6p$	$^{4}\mathrm{D}^{\circ}$	5/2	22804.25	10.7(1.0)	9.79	9.70		1.362	1.364
$5d^{2}(^{3}F)6p$	$^{4}\mathrm{D}^{\circ}$	7/2	23303.26	16.1(1.0)	10.32	9.91		1.178	1.417
$5d6s(^{3}D)6p$		7/2	23221.10	22.0(1.4)		f		1.078	-
$5d6s(^{3}D)6p$		5/2	23874.95	16.2(1.0)	12.57	17.10	$16(0.8)^c$	0.962	1.036
$4f5d(^{3}\mathrm{F}^{\circ})6s$	${}^{4}\mathrm{F}^{\circ}$	5/2	24507.87	21.9(1.0)	13.44	16.27	$14.5(1.5)^c$	1.158	1.185
$4f5d(^{3}\mathrm{F}^{\circ})6s$	${}^{4}\mathrm{F}^{\circ}$	7/2	25380.27	23.2(1.3)	11.74	12.40		1.228	1.227
$4f5d(^{3}\mathrm{F}^{\circ})6s$	${}^{4}\mathrm{F}^{\circ}$	9/2	25997.17	23.3(1.5)	11.91	12.78		1.319	1.325
$4f5d({}^{1}\mathrm{G}^{\circ})6s$		7/2	24409.68	15.7(0.7)	13.09	15.22	$16(0.6)^c$	1.161	1.201
$5d^2(^3\mathbf{P})6p$	$^{4}\mathrm{D}^{\circ}$	7/2	25083.36	21.1(0.9)	23.30	28.66		1.381	1.312
$5d^2(^3\mathbf{P})6p$	$^{2}P^{\circ}$	3/2	27225.26	17.1(0.9)	19.35	26.86		1.31	1.331
$5d^{2}(^{1}D)6p$		5/2	27393.04	14.1(0.6)	8.26	9.67^e		0.888	0.880
$4f5d(^{3}\mathrm{G}^{\circ})6s$	${}^{4}\mathrm{G}^{\circ}$	7/2	27455.31	21.6(1.6)	9.97	9.48		0.976	0.991
$4f5d(^{3}\mathrm{F}^{\circ})6s$	$^{2}\mathrm{F}^{\circ}$	5/2	27669.37	17.8(0.9)	24.50	18.56^{e}		0.88	0.887
$4f5d(^{3}\mathrm{F}^{\circ})6s$	$^{2}\mathrm{F}^{\circ}$	7/2	28543.08	21.7(1.2)		f		1.12	1.188
$5d6s(^{1}D)6p$		7/2	28039.45	13.0(0.9)		f		1.14	1.138
$5d6s(^{3}D)6p$		5/2	28506.41	7.9(0.3)	8.27	8.75	$9(1)^{c}$	1.20	1.142

^{*a*} Hese et al. [5] (Level-crossing method). ^{*b*} Bulos et al. [6] (Laser-induced fluorescence technique). ^{*c*} Penkin et al. [7] (Pulsedelectron delayed coincidence technique). ^{*d*} From Martin et al. [1]. ^{*e*} Strong mixing and nearly equal Landé factors are obtained for the two levels at 27 393.04 and 27 669.37 cm⁻¹. ^{*f*} For these levels, a strong mixing is observed and the identification of the levels is not sure even when considering the Landé factors.

results also from the gradual contraction of 4f with increasing Z, with the consequence that the 4f subshell is more and more buried inside the atomic core and interacts less and less strongly with the outer valence electrons when Z increases. Therefore, the lanthanide spectra appear somewhat less complex in the right part of the group than they are in the left part of the chart. In fact, the former spectra show contrasting strong and weak lines while the latter tend to be less contrasted regarding the intensities (see e.g. Wyart [19]; Cowan [20]), a fact which complicates the analysis of the spectra in this part of the periodic table, and of La I more specifically.

Despite of this complexity, we have performed some calculations of lifetime values in La I. The procedure followed for the calculations is similar to that adopted recently for obtaining theoretical branching fractions in doubly or singly ionised lanthanides (see e.g. Biémont et al. [21,22]). In a first calculation (calculation A), made with help of the Cowan's suite of computer codes (HFR method: see Cowan [20]), modified for taking care of corepolarisation effects (see e.g. Quinet et al. [23]), the following configurations were considered in the theoretical model: $5d6s^2 + 5d^26s + 5d^27s + 5d^26d + 5d^27d + 5d^3 + 5d66s^7s + 5d6p^2 + 5d66p^7p + 5d6d^2 + 5d6d7s + 5d6d7d +$

4f6s6p + 4f6s7p (even parity) and $5d6s6p + 5d6s7p + 6s^{2}6p + 5d^{2}6p + 5d^{2}7p + 5d6p7s + 5d6p6d + 5d6p7d + 4f6s^{2} + 4f5d^{2} + 4f6d^{2} + 4f5d6s + 4f6d7s + 4f5d6d + 4f5d7d$ (odd parity).

Core-polarisation (CP) effects, which are expected to be important in heavy neutral elements, were introduced in the calculations in a detailed way. More precisely, different cores were considered for each transition array and the dipole polarisabilities, α_d , and the cutoff radii, r_{c} , were obtained by considering the core as the "spectator" configuration, i.e. the configuration that remains when removing the active shells involved in the electronic transition. The numerical values of α_d were those computed by Fraga et al. [24]. As examples, when the transition array $5d6s^2 - 5d6s6p$ was considered, the "core" configuration was 5d (La III-type: $\alpha_d = 13.16$ a.u. and $r_c(5d) = 2.81$ a.u.); if the transition array $5d^26s - 5d^26p$ was considered, the "core" configuration was $5d^2$ (La IItype: $\alpha_d = 22.94$ a.u. and $r_c(5d) = 2.81$ a.u.) and when the transition array $5d6s^2 - 6s^26p$ was considered, the "core" configuration was $5p^6$ (La IV-type: $\alpha_d = 9.50$ a.u. and $r_c(5p) = 1.80$ a.u.).

The calculated eigenvalues of the Hamiltonian were optimized to the observed energy levels via a least-squares fitting procedure. In fact, all the levels included in the NIST compilation [1] below the ionisation limit, except those with unknown or dubious assignments (indicated with a question mark "?" in the tables) were included in the fitting procedure.

The scaling factors of the Slater parameters (F^k, G^k) and of configuration interaction integrals (R^k) , not optimized in the least-squares fitting, were chosen equal to 0.75 while the spin-orbit parameters were left at their ab initio values. This low value of the scaling factor is suggested by Cowan [20] for neutral heavy elements and has appeared to be adequate in recent calculations in neutral or lowly ionised lanthanides (see the relevant references in the database DREAM available on the web at the address: http://www.umh.ac.be/~astro/dream.shtml).

The theoretical HFR lifetime values, obtained in calculation A, are summarized in Table 2 (column 6) where they are compared with the measurements made in Lund (column 5). In most cases, the theoretical results are in satisfactory agreement with the experimental data; the discrepancies appear however rather large for some strongly perturbed levels. This could possibly result from the inadequacy of the model used for taking configurationinteraction effects into account. For checking this hypothesis, in an alternative calculation (calculation B), different configuration sets were used for obtaining the wavefunctions i.e. $5d6s^2 + 5d^26s + 5d6s6d + 5d^26d + 5d6p5f +$ $5d6p6f + 5d4f^2 + 5d^3 + 5d4f5f + 5d6p^2 + 5d6d^2 + 5d4f6f +$ $5d5f^2$ and $5d6s6p + 5d^26p + 5d6p6d + 4f5d^2 + 4f5d6d +$ $5d5f6d + 5d6d6f + 4f5d6s + 5d6s5f + 5d6s6f + 5d^25f +$ $5d^{2}6f$.

Core-polarisation effects were incorporated in the calculations following a procedure similar to that adopted in calculation A. The scaling factor adopted for the Slater and configuration-interaction integrals was 0.75 as in calculation A. The corresponding results are given in Table 2 under the heading B (column 7). We give also, in the last two columns of Table 2, a comparison between observed and calculated (calculation B) Landé factors, the agreement between the two sets of results giving more weight to the correspondence established between calculated and observed energy levels when a strong mixing is present.

As a rule, the calculated values (calculation B) do not differ substantially from the results of calculation A, except in a few cases where they have been substantially increased. It is observed that the levels for which the larger changes do appear are strongly perturbed, indicating that much larger configuration sets would probably be needed in order to obtain more stable lifetime values. This was not possible in the present paper in view of the limits imposed by the computer constraints.

In addition, it was checked that the way the corepolarisation effects were incorporated in the calculations had not a dramatic effect on the final values because a change by 10% of the static dipole polarisability of the core or of the cut-off radius (assuming here the same polarisation parameters for all the transition arrays) does not change the resulting lifetime values very much, the change being of the order of a few (<5)%, in agreement with previous findings in other ions of the same group of elements (see e.g. Ref. [25]).

4 Discussion of the results

As expected, the new measurement for the level at $18\,172.35\,\mathrm{cm}^{-1}$ agrees very well with the laser-induced fluorescence measurement of Bulos et al. [6]. An agreement, within the error bars, is also observed when comparing our experimental lifetime values with the delayed-coincidence results of Penkin et al. [7] for the levels situated at $18\,172.35$, $19\,379.40$, $23\,874.95$ and $24\,409.68\,\mathrm{cm}^{-1}$. A marginal difference (15%) does appear for $28\,506.41\,\mathrm{cm}^{-1}$ but a large discrepancy has to be noted for the level at $24\,507.87\,\mathrm{cm}^{-1}$. There is no obvious explanation for this last discrepancy. The two measurements of Hese et al. [4,5] by the level-crossing technique, for the levels at $18\,172.35$ and $19\,379.40\,\mathrm{cm}^{-1}$, appear systematically lower than all the other measurements indicating that these results are probably underestimated.

The rather poor agreement theory–experiment which is observed for some of the levels measured in the present work requires further consideration. Because the binding energies of 4f, 5d, 6s and 6p electrons in neutral lanthanides differ from each other by only a couple of electron-volts [20] whereas the spread of energies within a configuration reaches only a few eV, the low-lying configurations overlap each other to a very high degree. The consequence is that the density of energy levels becomes high even at energies as low as 2.0 eV above the ground level. The interpretation of experimental spectra and energy levels is consequently extremely difficult and, in view of the fragmentary knowledge of the levels or of the dubious assignment of some of them, the fitting procedure might be hazardous particularly for the odd-parity levels. This is confirmed by Ben Ahmed's [26] comments as reported by Martin et al. [1]. Her percentage compositions for the odd parity levels, deduced from a 5-configurations calculation $(5d6s6p + 6s^{2}6p + 5d^{2}6p + 4f6s^{2} + 4f5d6s)$ was preliminary and partly unsatisfactory since large deviations occurred for some levels in the region about $23\,000 \text{ cm}^{-1}$. The inclusion of additional configurations, particularly $4f5d^2$, would change the eigenvectors significantly, and possibly also some identifications. Except for the low levels of each configuration and the levels with high J, which are relatively few in number, the computed mixing of the basis wavefunctions is large and susceptible to render impossible a meaningful assignment of energy levels, besides parity and J value, and eventually configuration. The distinguishing characteristics of pure basis states are smeared out to such a degree that many levels for a given J and configuration tend to have similar properties including the numerical values of the Landé factors (which cannot be used as an identification tool). This can make some level assignments (and, consequently, the associated level-fitting procedure) unreliable and prevent the establishment of an unambiguous correspondence between the ab initio calculated and the experimentally determined levels.

In order to test the possible effects, on the lifetime values, of configurations not explicitly included in the above sets and possibly not mimicked by the scaling down of the Slater integrals, a third calculation with extended configuration sets (and consequently without inclusion of CP effects) was attempted. The configurations considered were the following: $5d6s^2 + 5d^26s + 5d^27s + 5d^3 + 5d6s7s + 5d^3 +$ $5d6s6d + 5d6s7d + 5d^26d + 5d6d7s + 5d6d^2 + 5d^27d + 4f6s6p + 5d6s7d + 5d^26d + 5d6d7s + 5d6d^2 + 5d^27d + 4f6s6p + 5d6d7s + 5$ $4f5d5f + 4f5d6f + 4f5d7f + 4f5d6p + 5d6p^2 + 5d6s6p +$ $5d6s7p + 5d^{2}6p + 5d^{2}7p + 5d4f6s + 5d4f7s + 5d5f6d +$ $5d^{2}4f + 5d^{2}5f + 5d^{2}6f + 5d6s5f + 5d6s6f + 5d6p6d + 5d6p7s.$ It is interesting to note that the corresponding lifetime values (not reported here) do not change significantly when compared with the results of calculations A and B (without core-polarisation effects included). This indicates that probably much larger sets of configurations would be necessary for each parity and that a renewed term analysis as well as an extension of the level system presently available are needed in order to refine the least-squares fitting procedure and get, in that way, a better and more realistic description of the real atom. This was, however, beyond our computer capabilities.

5 Conclusions

In the present paper, the first extensive set of accurate (uncertainty within 10%) lifetime measurements in La I has been obtained using selective laser excitation. The corresponding HFR calculations performed for obtaining a realistic description of the La I spectrum outline the complexity of this atomic system in relation with the sudden collapse of the 4f orbital occurring at Z = 57. They also emphasize the need for further laboratory investigations of this atom, concerning both term analysis and transition probability determination, in order to get the extensive set of data required by the astrophysicists. An additional difficulty of the HFR calculations results from the fact that the use of this approach is partly hindered by convergence problems appearing when calculating the wavefunctions of $4f^26s$, 4f5d6p and $4f6s^2$ configurations. New and extensive laboratory work concerning both line identification and branching fraction determination would be most welcome and even crucial at this prospect.

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