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Lifetimes along perturbed Rydberg series in neutral thallium

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Abstract

Radiative lifetimes of 15 Tl I levels belonging to the $6s^2ns\,^2S_{1/2}$ ($n = 7$–14) and $6s^2nd\,^2D_{3/2}$ Rydberg series ($n = 6$–12) have been measured using a time-resolved laser-induced fluorescence technique. All the measured levels have been excited from the ground state $6s^26p\,^2P^0_{1/2}$ (odd parity) with a single-step excitation process. The general perturbation of the ns series by the $6s6p^2$ configuration and the corresponding modification of the lifetimes are adequately reproduced by a theoretical model including core-polarization effects and combined with a least-squares fit to the observed energy levels. The general behaviour of the lifetime values for the $6s^2np$ odd levels along the Rydberg series is also well reproduced. The use of the multiconfiguration quantum defect theory has allowed us to obtain lifetime values along the $6s^2ns\,^2S_{1/2}$ series up to levels with $n = 31$.

1. Introduction

Thallium ($Z = 81$) has two stable isotopes, ^{203}Tl and ^{205}Tl, which are present in the solar system with relative abundances of 29.524% and 70.476%, respectively. In nucleosynthesis, these two isotopes are produced either by the r or the s process.

Up to now, Tl has been little investigated in astrophysical plasmas. This element has been identified, however, in sunspot spectra (Lambert *et al* 1969) and in the solar photosphere, the solar content ($A_{\rm Tl} = 0.90 \pm 0.20$, in the usual logarithmic scale) differing, however, considerably from the meteoritic abundance ($A_{\rm Tl} = 0.78 \pm 0.04$) (Asplund *et al* 2005). Tl is also, with arsenic, selenium and lead, one of the heaviest elements found in the interstellar gas (Cardelli and Ebbets 1993, Cardelli 1994), thanks to the observations carried out with the Goddard high-resolution spectrograph (GHRS) aboard the Hubble Space Telescope. These data allow us to investigate how the heavy elements chemically interact with interstellar dust

and they allow us to compare interstellar heavy elements at the time of formation of the solar system and in the current galactic epoch. Recently, Tl has been listed among the heavy elements which have been found to be strongly enhanced in the atmospheres of the chemically peculiar stars of the upper main sequence (Adelman *et al* 2004).

Tl has been the subject of many lifetime investigations (for a detailed list of references, see the NIST Atomic Database at the address http://physics.nist.gov/PhysRefData/Fvalbib/html/ref frm0.html) based on the use of a variety of experimental techniques. This includes a laser excited fluorescence technique in an inductively coupled plasma (Uchida *et al* 1983, 1984), the use of the after-glow of a pulsed rf discharge (Pickett and Anderson 1969), the phase-shift approach (Demtröder 1962, Cunningham and Link 1967), the hook method (Penkin and Shabanova 1963), the double resonance (Gallagher and Lurio 1964), the level crossing (Gallagher and Norton 1971), the Hanle effect (Gough and Griffiths 1977) and the beam-foil spectroscopy (Lindgård *et al* 1982, Andersen and Sorensen 1972, Andersen *et al* 1972).

Most of these investigations, however, are limited to a few low excitation energy levels ($n \leqslant 8$). In view of the technical difficulties involved when measuring long lifetimes, a very limited number of studies have been devoted to the investigation of the behaviour of atomic lifetimes along the Rydberg series up to higher ($\geqslant 8$) n values. They are due to Shimon and Erdevdi (1977) (ns, $n = 8$–10; nd, $n = 6$–9), James *et al* (1986) (np, $n = 7$–11) and Lindgård *et al* (1982) (nd, $n = 6$–8).

Tl is also of great interest for parity-nonconserving effects due to weak neutral currents. Accurate relativistic wavefunctions are required if the results of the measurements are to be compared with the predictions of different models of the weak neutral interaction (see, e.g. Neuffer and Commins (1977)). On the theoretical side, the neutral thallium atom has a rather simple electronic structure with a single valence electron and, consequently, has been the subject of several semi-empirical or theoretical investigations (see, e.g. Gruzdev (1966), Anderson *et al* (1967), Flambaum and Sushkov (1978), Migdalek and Baylis (1978, 1979a, 1979b) and Bardsley and Norcross (1980)). Comparison of theoretical and experimental lifetime values along the Rydberg series is very useful for refining the theoretical models because the long lifetime values are very sensitive to relativity and configuration interaction effects.

In fact, in Tl I, the simple structure of the type ns^2nl is perturbed by the existence of more complex configurations such as $nsnp^2$ which are caused by the break up of the filling of the ns subshell. Configuration interaction with these configurations has a strong influence on different atomic parameters such as fine-structure intervals, hyperfine-structure constants or oscillator strengths of electric dipole transitions. Similar perturbations are observed in many different situations like in the alkaline-earth atoms where perturbations of Rydberg series by isolated levels considerably modify the lifetime values (see, e.g. Bhatia *et al* (1981) and Aymar *et al* (1982)).

Migdalek and Baylis (1979a, 1979b) have demonstrated that correlation effects between valence and core electrons can be approximated by core-polarization corrections in relativistic single-configuration Hartree–Fock computations of oscillator strengths. This method has succeeded in removing large discrepancies between calculated and experimental oscillator strengths for $np^2P_J - (n+1)s^2S_J$ and $np^2P_J - nd^2D_J$ transitions in Ga I, In I and Tl I spectra. Bardsley and Norcross (1980) have discussed the importance of including, in the calculations, the core-polarization effects in both the model potential and the transition-matrix elements for computing accurate oscillator strengths and excited-state lifetime values. They have shown that the f values for transitions between low-lying states of Tl can be calculated with reasonable accuracy using a one-electron model provided that relativistic effects and

core-polarization are included in the model. A similar approach has been followed in the present work.

2. The HFR calculations

In order to allow for the inclusion, in the calculations, of the core-polarization effects, the relativistic Hartree–Fock method (HFR) (Cowan 1981) has been modified in a way previously described (Quinet *et al* 1999). The core–valence correlation can *a priori* be considered by opening the $6s^2$ and $5p^6$ shells but the number of possible interacting configurations to be introduced in the model is rapidly growing and computer limits quickly impose restrictions to that approach.

Instead, a core-polarization (CP) potential and a correction to the dipole operator, as described by Quinet *et al* (1999), were included in the model using the static dipole polarizability of the ionic core, α_d, and a cut-off radius, r_c, which corresponds to the expectation value of r for the outermost core orbital (6s) as calculated with the HFR approach. For the first parameter, we used dipole polarizabilities tabulated by Fraga *et al* (1976) for the ionic cores as calculated in an approximation based on HF wavefunctions.

Two different calculations were performed: in the first one (calculation A), the configurations $np\,(n = 6\text{–}15) + nf\,(n = 5\text{–}15) + nh\,(n = 6\text{–}15) + 6p7s + 6p6d + 6p7d$ (odd parity) and $ns\,(n = 7\text{–}15) + nd\,(n = 6\text{–}15) + ng\,(n = 5\text{–}15) + 6s6p^2 + 6s6d^2 + 6s5f^2 + 6s6f^2 + 6p7p + 6p5f + 6p6f + 6p7f$ (even parity) were considered in the calculations. We adopted α_d(Tl III) = 2.37 (Å^3), i.e. $\alpha_d\left(a_0^3\right) = 15.994$ and $r_c = 2.347a_0$, which corresponds to the outermost orbital (6s) in the $5d^{10}\,6s$ configuration. In a second calculation (calculation B), the wavefunctions were calculated with the HFR approach including the configurations $np\,(n = 6\text{–}15) + nf\,(n = 5\text{–}15) + nh\,(n = 6\text{–}15)$ (odd parity) and $ns\,(n = 7\text{–}15) + nd\,(n = 6\text{–}15) + ng\,(n = 5\text{–}15) + 6s6p^2$ (even parity). We adopted α_d(Tl II) = 5.53 which corresponds to $\alpha_d\left(a_0^3\right) = 37.318$ and r_c was chosen equal to $2.480a_0$, considering the 6s orbital within the configuration $5d^{10}\,6s^2$. This value of the polarizability was also adopted by Migdalek and Baylis (1979c). It is, however, substantially larger than the result quoted by Bardsley and Norcross (1980), i.e. $\alpha_d = 17.0$ au. The lifetime values obtained with calculation B were in closer agreement with the experimental measurements than the calculation A (not reproduced here) and, consequently, they were adopted as the final results. Migdalek and Baylis (1978, 1979c) compared two versions of the computations including core-polarization effects. In the first one, the static dipole polarizability of the core was adjusted for the ground state of the atom to bring the theoretical energy of the valence electron to within 0.1% of the experimental ionization energy. In the second one, which was found to be superior, the numerical values of α_d were taken directly from Fraga *et al* (1976). We followed this second procedure here. It should be emphasized that the polarizability obtained by Fraga *et al* (1976) agrees quite well with the numerical values as calculated by Flambaum and Sushkov (1978) (i.e. $\alpha_d\left(a_0^3\right) = 37.3$).

In addition to the inclusion in the calculation of the core-polarization effects, a least-squares fitting procedure has been considered based on the energy levels listed by Moore (1958). In this compilation, energy levels along the Rydberg series have been listed up to $n = 20\,(ns)$, $n = 16\,(np)$, $n = 23\,(nd)$ and $n = 8\,(nf)$. The level values in Moore's (1958) tables are taken basically from old analyses carried out by Paschen and Götze (1922) and Fowler (1922) with further additions taken from Beutler and Demeter (1934), Clearman (1952) and Meggers and Murphy (1952). In the NIST compilation (Moore 1958), five levels belonging to the configuration $6s6p^2$ are reported, two of them appearing above the first ionization stage. We did exclude from the fitting procedure the level at 62 000 cm^{-1} that it

was not possible to fit accurately. The level observed at 45 220 cm^{-1} appears strongly mixed with the $10s\,^2S_{1/2}$ level, the percentage composition reaching only 46.9%.

The theoretical lifetime values as obtained in the present work (calculation B) are reported in table 1 where they are compared with the experiment and with previous theoretical and experimental results.

3. Lifetime measurements

In the present experiment, radiative lifetimes of 15 Tl I Rydberg levels, belonging to the $6s^2ns\,^2S_{1/2}$ (n = 7–14) and $6s^2nd\,^2D_{3/2}$ series (n = 6–12), were measured using a time-resolved laser-induced fluorescence (LIF) technique (Biémont *et al* 2004, Xu *et al* 2004). All the measured levels were excited from the ground state $6s^26p\,^2P^0_{1/2}$ (odd parity) with a single-step excitation process.

Free thallium atoms were produced in a small plasma by laser ablation. A solid target of $Tl_2\ O_3$ (99.99%), rotating in a vacuum chamber with 10^{-6} to 10^{-5} mbar, was irradiated perpendicularly by a 532 nm laser pulse, emitted from an Nd:YAG laser (Continuum Surelite) with 10 ns pulse duration. The pulse energy used was normally in the range of 2–10 mJ and caused a small plasma expanding from the target. After the interaction between the ions and electrons, the neutral thallium atoms were produced and moved into the interaction zone about 10 mm above the target.

The excitation laser had a pulse duration of about 1 ns, which was produced by sending a primary pumping laser pulse, emitted from a seeder injected Nd:YAG laser (Continuum NY-82) with 8 ns pulse duration at 532 nm, to a temporal compressor, which is based on the stimulated Brillouin scattering (SBS) technique in water. These compressed pulses were used to pump a dye laser (Continuum Nd-60) operated with the dye DCM to give a tunable radiation. The required excitation wavelengths were obtained using different nonlinear optical techniques, such as frequency upconversion in crystals and stimulated Raman components in Raman scattering cell with hydrogen gas at 10 bar. The description of experimental apparatus can be found elsewhere (Biémont *et al* 2004, Xu *et al* 2004) where more details are given.

4. Discussion of the results

The radiative lifetimes measured (column 4) and calculated (column 6) in the present work are compared in table 1. Previous experimental and theoretical results are also reported in the same table for comparison. For $6s^27s\,^2S_{1/2}$ and $6s^26d\,^2D_{3/2}$ levels, the LIF lifetimes were obtained by deconvolution of the time-resolved signal with the excitation pulse, as shown in figure 1. For the other levels, the lifetimes were evaluated with exponential fitting procedures, as shown in figure 2.

For $6s^2ns\,^2S_{1/2}$ levels (n = 7–10), some scatter is observed among the different experimental results. The LIF lifetime value of the $6s^27s\,^2S_{1/2}$ state is in a good agreement with the previous data obtained by Shimon and Erdevdi (1977), Lindgård *et al* (1982), Gallagher and Lurio (1964) and Cunningham and Link (1967). For $8s\,^2S$, the different experimental results agree well. Larger discrepancies are observed for $9s\,^2S$ and $10s\,^2S$ between the LIF results and the previous data. For the whole $ns\,^2S$ series, the LIF results, however, agree quite well with the HFR theoretical results as calculated in the present work, even for the $10s\,^2S_{1/2}$ level which is strongly perturbed, indicating that the present theoretical model is very satisfying particularly for describing the perturbation affecting the whole series.

Table 1. Measured and calculated radiative lifetimes (in ns) for Tl I Rydberg series.

E (cm^{-1})	Configuration	J	Expt[a]	Expt (Prev.)	Theory[a]	Theory (Prev.)[b]
				*n*s series		
26 477.5	$6s^2 7s\,^2S$	1/2	7.3 ± 0.4	7.4 ± 0.5[c], 7.3 ± 0.3[d], 6.9 ± 1.0[e], 7.7 ± 0.5[f], 8.2 ± 0.6[g], 7.4 ± 0.3[h], 7.4 ± 0.2[i], 7.6 ± 0.2[h,j], 6.3 ± 0.7[e], 8.5 ± 0.3[k], 7.55 ± 0.08[l]	6.79	7.17
38 745.9	$6s^2 8s\,^2S$	1/2	25 ± 2	20 ± 3[c], 22 ± 2[e], 19 ± 2[e], 23 ± 4[f]	21.59	20.9
43 166.2	$6s^2 9s\,^2S$	1/2	54 ± 4	43 ± 4[c], 40 ± 10[e]	57.03	46.0
45 296.8	$6s^2 10s\,^2S$	1/2	50 ± 4	31 ± 3[c]	53.32	86.1
46 456.9	$6s^2 11s\,^2S$	1/2	145 ± 10		120.3	
47 178.9	$6s^2 12s\,^2S$	1/2	225 ± 20		210.4	
47 654.7	$6s^2 13s\,^2S$	1/2	310 ± 30		329.4	
47 983.2	$6s^2 14s\,^2S$	1/2	410 ± 50		481.1	
				*n*d series		
36 117.9	$6s^2 6d\,^2D$	3/2	8.5 ± 0.5	6.9 ± 0.5[c], 6.1 ± 0.7[e], 6.8 ± 0.5[f]	5.90	7.04
36 199.9		5/2		7.2 ± 0.6[c], 6.5 ± 0.7[e], 6.8 ± 0.4[m]	11.09	8.06
42 011.4	$6s^2 7d\,^2D$	3/2	20.5 ± 1.5	16.0 ± 1.3[c], 13 ± 2[e], 16 ± 4[f]	16.48	17.5
42 049.0		5/2		19.8 ± 1.5[c], 16 ± 3[e], 19 ± 4[f]	27.25	20.5
44 672.6	$6s^2 8d\,^2D$	3/2	45 ± 5	34 ± 3[c], 27 ± 5[e]	37.12	35.0
44 692.7		5/2		44.0 ± 4[c], 26 ± 5[e], 50 ± 10[f]	57.93	41.3
46 098.5	$6s^2 9d\,^2D$	3/2	56 ± 6	75 ± 7[c]	71.79	61.6
46 110.3		5/2			108.5	72.8
46 949.9	$6s^2 10d\,^2D$	3/2	81 ± 9		125.0	
46 958.0		5/2			185.3	
47 499.8	$6s^2 11d\,^2D$	3/2	130 ± 20		202.2	
47 504.1		5/2			296.0	
47 876.0	$6s^2 12d\,^2D$	3/2	210 ± 30		311.1	
47 876.0		5/2			452.3	
				*n*p series		
34 159.9	$6s^2 7p\,^2P^0$	1/2		63.1 ± 1.7[n], 58.5[o], 61.9[p]	59.36	61.0
35 161.1	$6s^2 7p\,^2P^0$	3/2		48.6 ± 1.3[n], 42.2[o], 48.4[p]	41.11	47.4
41 368.1	$6s^2 8p\,^2P^0$	1/2		184.1 ± 4.4[n]	157.4	185.0
41 470.8	$6s^2 8p\,^2P^0$	3/2		127.7 ± 4.9[n], 108.7[o], 123 ± 7[q]	125.3	130.0
44 380.9	$6s^2 9p\,^2P^0$	1/2		391.1 ± 21.8[n]	323.1	380.0
44 562.5	$6s^2 9p\,^2P^0$	3/2		273.6 ± 13.5[n], 16.4 ± 1.5[c], 265 ± 14[q]	277.2	267.0
45 939.3	$6s^2 10p\,^2P^0$	1/2		656.8 ± 14.5[n]	570.6	
46 043.6	$6s^2 10p\,^2P^0$	3/2		480.8 ± 31.6[n]	509.4	

Table 1. (Continued.)

E (cm^{-1})	Configuration	J	Expt[a]	Expt (Prev.)	Theory[a]	Theory (Prev.)[b]
46 853.8	$6s^2 11p\,^2P^0$	1/2		991.1 ± 50.8[n]	929.6	
46 917.1	$6s^2 11p\,^2P^0$	3/2		725.5 ± 28.8[n]	854.2	
			$6s6p^2\,^4P$			
45 220	$6s6p^2\,^4P$	1/2		40 ± 10[e]	37.55	

[a] This work.
[b] Bardsley and Norcross (1980): theory.
[c] Shimon and Erdevdi (1977): delayed coincidence method (electron beam).
[d] Harvey *et al* (1977): laser spectroscopy.
[e] Lindgård *et al* (1982): beam-foil method.
[f] Andersen and Sorensen (1972): beam-foil method.
[g] Penkin and Shabanova (1963): hook method.
[h] Gallagher and Lurio (1964): double resonance.
[i] Gallagher and Norton (1971): level crossing.
[j] Cunningham and Link (1967): phase-shift method.
[k] Demtröder (1962): phase-shift method.
[l] Hsieh and Baird (1972): level crossing.
[m] Gough and Griffiths (1977): Hanle effect.
[n] James *et al* (1986): atomic beam with two-photon excitation and fluorescence decay.
[o] Hunter *et al* (1982): Hanle effect.
[p] James *et al* (1985): two-photon resonant excitation.
[q] Hermann *et al* (1988): level-crossing spectroscopy.

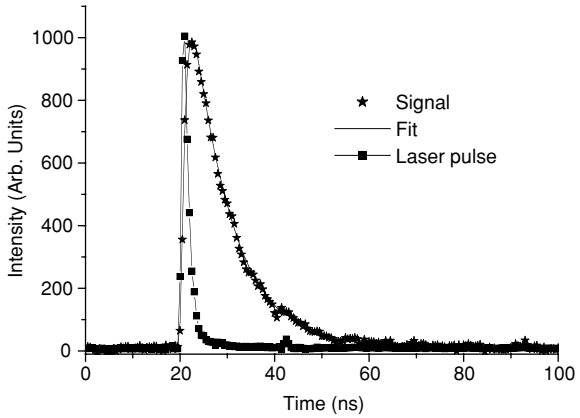

Figure 1. Experimental decay curve for the $6s^2 7s$ state of Tl I and the recorded excitation laser pulse.

The lifetime values for the $6s^2 nd\,^2D_{3/2}$ states measured in this work are systematically larger than the other measurements. They also appear larger than the HFR results of the present work for the first members ($n = 6$–8) of the series, the contrary appearing for the higher members. The influence of possible Zeeman quantum beat effects on the D-level measurements was eliminated by applying an external magnetic field of sufficient strength to wash out the oscillations.

Several additional effects, such as collisions and flight-out-of-view effects, which can generate errors in experimental lifetimes have also been considered. In the measurements,

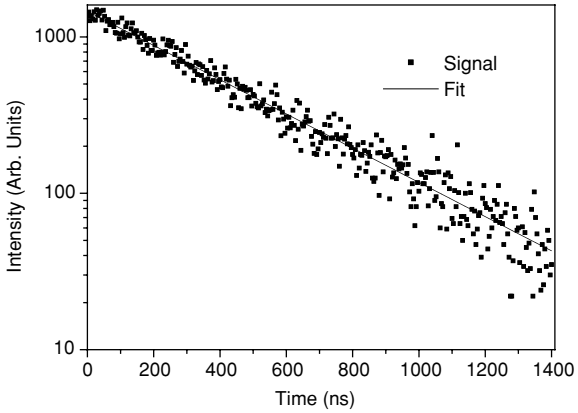

Figure 2. Experimental decay curve for the $6s^2 14s$ state of Tl I. The exponential fit gives a lifetime of 410 ± 50 ns.

attempts were made to eliminate these effects by adjusting the experimental parameters. To make sure that the experimental lifetimes were not affected by collisions and radiation trapping, the intensity of the ablation pulse and the delay time were changed, that is, the atomic density and temperature were modified. The signal intensities were varied, but the lifetime values were found to be constant, which implied that collisional quenching and radiation trapping effects were negligible. Different neutral density filters, inserted in the exciting light path, were used to avoid the saturation effect. An important aspect in lifetime measurements consists indeed in avoiding flight-out-of-view effects, especially when the measured lifetimes are long. In the present measurements, the entrance slit of the monochromator was put perpendicularly and parallel, respectively, to the direction of the plasma movement. Long delay times (10 μs) between the ablation and excitation lasers were also adopted to minimize the possible flight-out-of-view effects. The error bars of the values in table 1 were determined from statistical spread of the different curve recordings.

For the np Rydberg series, not considered experimentally in the present work, the HFR theoretical lifetimes agree in a satisfying way with the previous experimental results, particularly those published by James *et al* (1985, 1986) and Hunter *et al* (1982). A good agreement with the theory of Bardsley and Norcross (1980) is also observed.

An additional argument in favour of the present theoretical model is provided by the lifetime value calculated for the strongly perturbed $6s6p^2\,{}^4P_{1/2}$ level. The HFR value ($\tau = 37.55$ ns; see last value in table 1) is found in excellent agreement (in fact, within the error bars) with the measurement by Lindgård *et al* (1982), i.e. $\tau = 40 \pm 10$ ns.

The dependence of the experimentally measured Rydberg lifetimes as a function of the effective principal quantum number, n^*, is shown in figure 3 in a double-logarithmic representation. The solid line denotes the dependence of lifetime versus n^* with the slope of 3. These curves show some irregularities due to the configuration interaction as expected from the theoretical predictions.

The absorption oscillator strengths and decay transition rates for the sharp, diffuse and principal series of Tl I are given in table 2 where they are compared with the previous theoretical results due to Bardsley and Norcross (1980) and with the experimental measurements (arc plasma) of Alonso-Medina (1997). A reasonably good agreement is generally observed between the three sets of results.

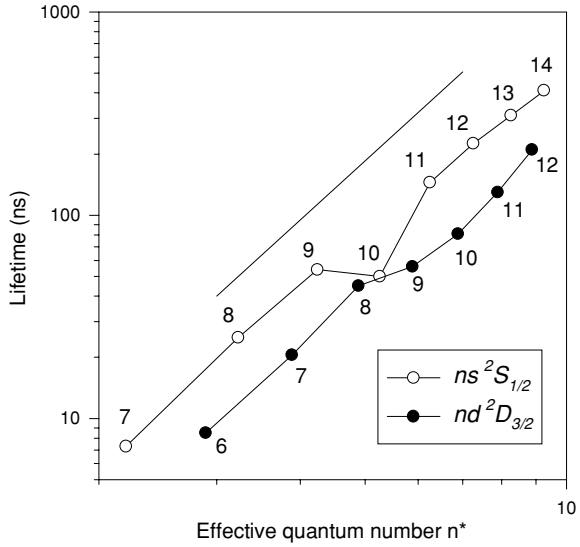

Figure 3. Lifetimes plotted versus the effective quantum number n^* for the $ns\,^2S$ and $nd\,^2D$ series of Tl I. The principal quantum numbers of the levels are indicated in the figure.

5. The MQDT approach

Multichannel quantum defect theory (MQDT), first proposed by Seaton (1966) and reformulated by Fano (1975), has been extensively and successfully applied to the description of perturbed Rydberg series in atoms (Armstrong *et al* 1977, Aymar and Robaux 1979, Aymar *et al* 1980, Hasegawa and Suzuki 1996, Dai *et al* 2002). MQDT wavefunctions derived from experimentally determined level energies are useful for predicting other spectroscopic properties such as radiative lifetimes, Landé factors and hyperfine structures. Details about theoretical methods and formulae for lifetime calculation of perturbed Rydberg states with MQDT wavefunctions can be found in Aymar *et al* (1981) and Dai *et al* (2002).

On the basis of the energy level data known to date (Moore 1958), it appears that the $6s^2ns\,^2S_{1/2}$ series of Tl I is perturbed by the $6s6p^2\,^4P_{1/2}$ interacting level belonging to the $6s6pnp\,^4P_{1/2}$ channel. So our MQDT analysis includes two collision channels: $6s^2ns\,^2S_{1/2}$ and $6s6pnp\,^4P_{1/2}$, the former converging to the ionization limit $6s^2\,^1S_0$ (49 264.2 cm^{-1}) and the latter to the ionization limit $6s6p\,^3P^0_2$ (61 725 cm^{-1}). Besides the perturber $6s6p^2\,^4P_{1/2}$, 14 experimental $6s^2ns\,^2S_{1/2}$ ($n = 7$–20) Rydberg levels, in the energy range from 26 477.5 cm^{-1} up to 48 796.2 cm^{-1} (Moore 1958), were used in the MQDT calculation. By fitting the theoretical values of the energy levels to the experimental ones, the optimal MQDT parameters and wavefunctions were obtained. The theoretical energy levels (up to $n = 31$), as deduced for the investigated series, are shown in table 3 (column 3).

Under the electric dipole approximation, the formula for obtaining the radiative decay rate of the ith level of the $6s^2ns\,^2S_{1/2}$ Rydberg series perturbed by $6s6p^2\,^4P_{1/2}$ can be expressed as

$$\Gamma_i = \gamma_1 \frac{{Z_1^i}^2}{n_i^{*}} + \gamma_2 {Z_2^i}^2, \tag{1}$$

where γ_1 and γ_2 are the radiative rate parameters corresponding to the $6s^2ns\,^2S_{1/2}$ and $6s6pnp\,^4P_{1/2}$ channels, respectively. n_i^* is the effective quantum number relatively to the

Table 2. Absorption oscillator strengths and decay transition rates for the sharp, diffuse and principal series of Tl I and comparison with previous results.

Transition	J–J	log gf	gA (this work)	gA (a)	gA (b)
$6\,^2P^0$–$7\,^2S$	1/2–1/2	−0.43	1.723(8)	1.266(8)	
$6\,^2P^0$–$8\,^2S$	1/2–1/2	−1.46	3.472(7)	3.780(7)	
$6\,^2P^0$–$9\,^2S$	1/2–1/2	−2.05	1.105(7)	1.666(7)	
$6\,^2P^0$–$10\,^2S$	1/2–1/2	−2.76	2.367(6)	8.820(6)	2.700(7)
$6\,^2P^0$–$11\,^2S$	1/2–1/2	−2.34	6.535(6)		5.400(6)
$6\,^2P^0$–$12\,^2S$	1/2–1/2	−2.63	3.507(6)		3.400(6)
$6\,^2P^0$–$13\,^2S$	1/2–1/2	−2.84	2.215(6)		2.200(6)
$6\,^2P^0$–$14\,^2S$	1/2–1/2	−3.01	1.507(6)		1.020(6)
$6\,^2P^0$–$15\,^2S$	1/2–1/2	−3.16	1.074(6)		
$6\,^2P^0$–$7\,^2S$	3/2–1/2	−0.28	1.222(8)	1.528(8)	
$6\,^2P^0$–$8\,^2S$	3/2–1/2	−1.24	3.707(7)	4.000(7)	
$6\,^2P^0$–$9\,^2S$	3/2–1/2	−1.77	1.404(7)	1.724(7)	
$6\,^2P^0$–$10\,^2S$	3/2–1/2	−4.49[a]	3.007(4)[a]	9.040(6)	1.200(7)
$6\,^2P^0$–$11\,^2S$	3/2–1/2	−2.18	6.624 (6)		5.600(6)
$6\,^2P^0$–$12\,^2S$	3/2–1/2	−2.43	3.814(6)		3.400(6)
$6\,^2P^0$–$13\,^2S$	3/2–1/2	−2.63	2.480(6)		2.000(6)
$6\,^2P^0$–$14\,^2S$	3/2–1/2	−2.80	1.716(6)		1.080(6)
$6\,^2P^0$–$15\,^2S$	3/2–1/2	−2.95	1.236(6)		
$6\,^2P^0$–$6\,^2D$	1/2–3/2	−0.15	6.152(8)	4.840(8)	
$6\,^2P^0$–$7\,^2D$	1/2–3/2	−0.81	1.823(8)	1.748(8)	
$6\,^2P^0$–$8\,^2D$	1/2–3/2	−1.27	7.197(7)	8.200(7)	
$6\,^2P^0$–$9\,^2D$	1/2–3/2	−1.62	3.435(7)	4.480(7)	4.240(7)
$6\,^2P^0$–$10\,^2D$	1/2–3/2	−1.90	1.859(7)		2.160(7)
$6\,^2P^0$–$11\,^2D$	1/2–3/2	−2.14	1.099(7)		1.040(7)
$6\,^2P^0$–$12\,^2D$	1/2–3/2	−2.34	6.921(6)		
$6\,^2P^0$–$13\,^2D$	1/2–3/2	−2.53	4.533(6)		
$6\,^2P^0$–$14\,^2D$	1/2–3/2	−2.71	3.052(6)		
$6\,^2P^0$–$15\,^2D$	1/2–3/2	−2.89	2.023(6)		
$6\,^2P^0$–$6\,^2D$	3/2–3/2	−0.94	6.112(7)	8.400(7)	
$6\,^2P^0$–$7\,^2D$	3/2–3/2	−1.57	2.096(7)	2.724(7)	
$6\,^2P^0$–$8\,^2D$	3/2–3/2	−2.01	8.834(6)	1.220(7)	
$6\,^2P^0$–$9\,^2D$	3/2–3/2	−2.35	4.369(6)	6.560(6)	8.000(6)
$6\,^2P^0$–$10\,^2D$	3/2–3/2	−2.63	2.411(6)		3.120(6)
$6\,^2P^0$–$11\,^2D$	3/2–3/2	−2.86	1.439(6)		
$6\,^2P^0$–$12\,^2D$	3/2–3/2	−3.07	9.097(5)		
$6\,^2P^0$–$13\,^2D$	3/2–3/2	−3.26	5.956(5)		
$6\,^2P^0$–$14\,^2D$	3/2–3/2	−3.44	3.998(5)		
$6\,^2P^0$–$15\,^2D$	3/2–3/2	−3.62	2.638(5)		
$6\,^2P^0$–$6\,^2D$	3/2–5/2	0.00	5.407(8)	7.440(8)	
$6\,^2P^0$–$7\,^2D$	3/2–5/2	−0.64	1.778(8)	2.472(8)	
$6\,^2P^0$–$8\,^2D$	3/2–5/2	−1.10	7.217(7)	1.122(8)	
$6\,^2P^0$–$9\,^2D$	3/2–5/2	−1.45	3.464(7)	6.060(7)	6.720(7)
$6\,^2P^0$–$10\,^2D$	3/2–5/2	−1.74	1.873(7)		4.440(7)
$6\,^2P^0$–$11\,^2D$	3/2–5/2	−1.98	1.102(7)		
$6\,^2P^0$–$12\,^2D$	3/2–5/2	−2.19	6.908(6)		6.600(6)

Table 2. (Continued.)

Transition	J–J	log gf	gA (this work)	gA (a)	gA (b)
$6\,^2P^0$–$13\,^2D$	3/2–5/2	−2.38	4.517(6)		
$6\,^2P^0$–$14\,^2D$	3/2–5/2	−2.56	3.037(6)		
$6\,^2P^0$–$15\,^2D$	3/2–5/2	−2.74	2.010(6)		

[a] Cancellation effects present. $a(b)$ is written for $a \times 10^b$.
(a) From Bardsley and Norcross (1980).
(b) From Alonso-Medina (1997).

Table 3. MQDT lifetimes for Tl I $6s^2ns$ series ($J = 1/2$) by fitting experimental lifetimes (ns) and HFR theoretical lifetimes (ns), respectively.

n	E^{a}_{expt}	E_{MQDT}	τ^{b}_{expt}	τ^{c}_{MQDT}	τ^{b}_{HFR}	τ^{d}_{MQDT}
7	26 477.5	26 475.3	7.3 ± 0.4	5.9	6.8	6.5
8	38 745.9	38 736.6	25 ± 2	18.4	21.6	20.1
9	43 166.2	43 160.5	54 ± 4	41.4	57.0	45.1
	45 220.0 (perturber)	45 161.2		32.3		31.9
10	45 296.8	45 326.9	50 ± 4	50.9	53.3	52.1
11	46 456.9	46 458.9	145 ± 10	131.5	120.3	143.2
12	47 178.9	47 179.6	225 ± 20	206.2	210.4	224.6
13	47 654.7	47 654.4	310 ± 30	304.1	329.4	331.4
14	47 983.2	47 983.7	410 ± 50	428.8	481.1	467.2
15	48 223.2	48 221.3		583.4		635.8
16	48 399.5	48 398.4		771.4		840.6
17	48 534.8	48 533.9		995.9		1085.2
18	48 639.0	48 640.0		1260.1		1373.2
19	48 726.2	48 724.5		1567.4		1708.1
20	48 796.2	48 792.9		1920.9		2093.3
21		48 849.1		2323.9		2532.5
22		48 895.8		2779.7		3029.2
23		48 935.1		3291.5		3587.0
24		48 968.4		3862.4		4209.2
25		48 996.8		4496.0		4899.6
26		49 021.4		5195.2		5661.6
27		49 042.7		5963.3		6498.7
28		49 061.4		6803.8		7414.7
29		49 077.7		7719.7		8412.8
30		49 092.2		8714.1		9496.4
31		49 105.1		9790.7		10 669.7

[a] From Moore (1958).
[b] This work.
[c] Obtained from a fit of the experimental lifetimes.
[d] Obtained from a fit of the HFR lifetimes.

first limit while $Z^i_{1,2}$ are the admixture coefficients of the first and second channels. The lifetime τ_i of the level i can be calculated by

$$\tau_i = \frac{1}{\Gamma_i}. \tag{2}$$

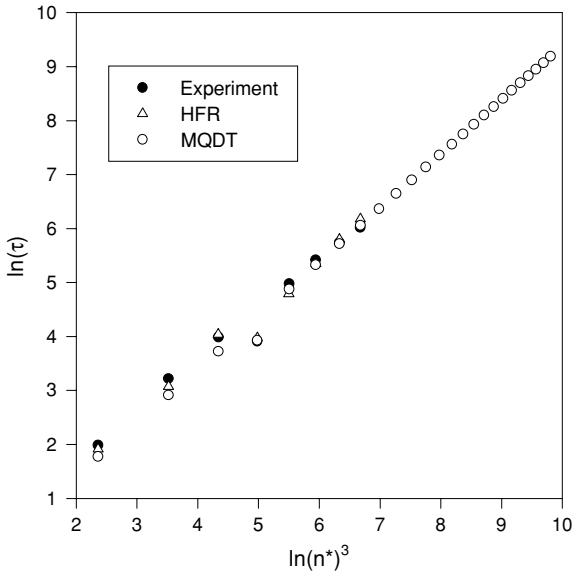

Figure 4. Comparison between MQDT, HFR and experimental lifetimes, τ (in ns), along the $6s^2ns$ Rydberg series of Tl I. n^* is the effective principal quantum number.

Using the MQDT admixture coefficients and fitting the theoretical lifetimes to the eight experimental lifetimes of the $6s^2ns\,^2S_{1/2}$ ($n = 7$–14) Rydberg states measured in this work, we have obtained the following radiative rate parameters: $\gamma_1 = 18.465 \times 10^8$ s^{-1}, $\gamma_2 = 0.381 \times 10^8$ s^{-1}. On the basis of these two parameters, MQDT theoretical lifetimes of the $6s^2ns\,^2S_{1/2}$ series ($n = 7$–31) and of the $6s6p^2\,^4P_{1/2}$ level have been calculated and the results are shown in table 3 (column 5). Moreover, we attempted to obtain MQDT lifetimes by fitting the HFR theoretical results obtained in the present work. The corresponding results are presented in table 3 (column 7). In this calculation, we deduced $\gamma_1 = 16.940 \times 10^8$ s^{-1} and $\gamma_2 = 0.391 \times 10^8$ s^{-1}. The root mean square (rms) deviation between the MQDT and experimental lifetimes is 3.61 ns while the rms deviation between the MQDT and HFR lifetimes is 3.37 ns.

Comparisons between MQDT, HFR and experimental lifetimes are presented in figure 4. As seen from this figure, the MQDT lifetimes are in good agreement with the experimental and HFR data except for the few levels near the perturber. Since the configuration admixture of the $6s6p^2\,^4P_{1/2}$ perturber into the studied Rydberg series is not large, the radiative lifetimes in the series are basically determined by the ratio

$$\frac{n^{*3}}{\gamma_1}. \tag{3}$$

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