Radiative lifetimes of highly excited states in rubidium

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Résumé. — Nous avons mesuré les durées de vie radiative d'états S et D très excités ($9 \leqslant n \leqslant 18$) du rubidium. Les résultats obtenus ainsi que ceux déjà publiés pour les états P et F sont en bon accord avec les valeurs théoriques calculées par la méthode de Bates-Damgaard.

Abstract. — Measurements have been made of the natural radiative lifetimes of highly excited rubidium ($9 \leqslant n \leqslant 18$) S and D states. These results, as well as those already published concerning P and F states, agree well with the theoretical lifetimes calculated by the Bates-Damgaard method.

In the past few years, measurements of the radiative lifetimes of highly excited states of alkali atoms have been reported [1-3]. These results would clearly be of interest for physicists working in atomic physics, plasma physics, astrophysics.

Moreover they allow interesting comparisons with theoretical calculations. We have already published measurements of the radiative lifetimes for some Rydberg P and F states in rubidium [4, 5]. In order to obtain a consistent set of lifetime values for a given alkali atom, we present here measurements concerning the nS and nD states of rubidium ($9 \leqslant n \leqslant 18$). Comparison is given between the obtained experimental values and the theoretical ones calculated by using a modified Bates-Damgaard Method [6, 7] (here after refered as B.D.).

We use the classical method of time resolved laser induced fluorescence [4]. Rubidium atoms contained in a cell are excited in a given high-lying nS or nD state by stepwise pulsed excitation via the first resonant 5P state. For convenience the $5P_{3/2}$ state is chosen as the intermediate state for the excitation of the nS levels, the observation of the population decay being made on the $n\text{S} \rightarrow 5P_{1/2}$ line. The nD states are produced by using light pulses whose wavelengths correspond either, to the $5P_{1/2} \rightarrow nD_{3/2}$ transitions for $n \leqslant 13$, or to the $5P_{3/2} \rightarrow nD_{3/2,5/2}$ transitions for $n > 13$; the population decay is observed on these same lines. Such a choice is determined by the fact that we are also interested in the study of some collisional properties of the nD states (results concerning the collisional properties of both the S and D levels will be published elsewhere [8]). Thus our reported lifetimes refer to the $nD_{3/2}$ states for $n \leqslant 13$ and to the nD states, without allowance for fine structure, for $n > 13$. This is of no importance since the lifetimes corresponding to the two fine structure levels are expected to be very close, for a given n value.

A high-speed photon counting system allows the determination of the decay of the excited state population as a function of time for a given rubidium pressure. The extrapolation to zero rubidium pressure yields the natural radiative lifetime. The experimental set-up, shown in figure 1, has been already widely

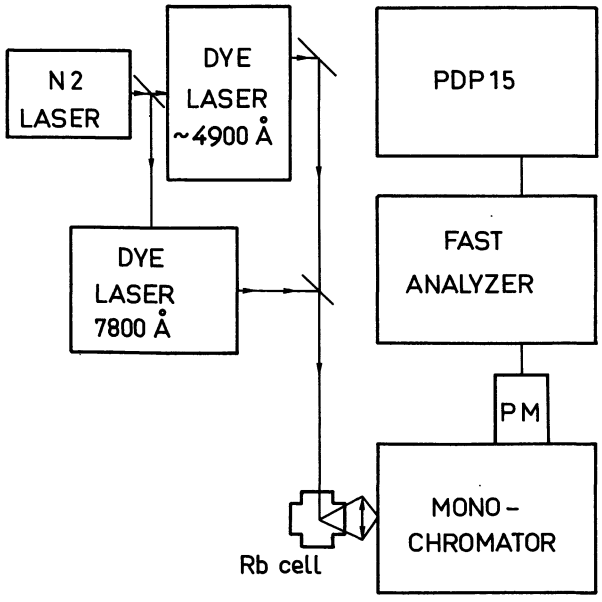

Fig. 1. — Experimental set-up.

described and needs no further explanations [5]. All the experiments were performed at $T = 520$ K. The working rubidium pressure lies in the 3×10^{-6} to 9×10^{-4} torr range. The results shown in table I have been corrected for pile-up and thermal escape effects. Great care has been taken to avoid parasitic effects due to superradiance which might be important for Rydberg states [9]. Other causes of error have already been widely discussed [5, 10]. The main uncertainty comes from the determination of the rubidium pressure which was determined from both the temperature of the side arm containing the liquid rubidium and from absorption measurements of the resonance rubidium line [4, 5].

Table I. — *Experimental lifetimes* τ_{exp} *for rubidium excited states. The* τ_{th} *values are from* [7]. *See text for* τ_{cal} *values. All values in* 10^{-9} s.

Level	τ_{exp}	τ_{th}	τ_{cal}
9S	245 ± 50	274	256
12S	770 ± 150	887	732
14S	1 260 ± 250	1 600	1 230
16S	2 190 ± 500	2 620	1 890
17S	2 600 ± 600	3 260	2 290
18S	3 300 ± 700	4 000	2 730
9D	565 ± 120	800	705
10D	720 ± 120	1 070	915
11D	975 ± 200	1 410	1 170
12D	1 250 ± 300	1 830	1 470
13D	1 400 ± 300	2 330	1 830
15D	3 740 ± 700	3 610	2 670
18D	5 300 ± 1 100	6 320	4 310

The B. D. method is the simplest for obtaining theoretical determinations of the radiative lifetimes for Rydberg states. We have already reported such extensive calculations in the case of the alkali atoms [7]. Comparison is given, for all the measured S, P, D and F levels, between theoretical τ_{th} and experimental τ_{exp} values in figure 2. For the S, P and D levels the τ_{exp} values are observed to be somewhat smaller than the τ_{th} values. This discrepancy can be partially removed by taking into account the interaction of the black-body radiation with the highly excited atoms [11]. In order to account for this effect we have computed the lifetime values τ_{cal}, which depend now on the temperature of the experimental cell, using the equation

$$\frac{1}{\tau_{cal}} = \frac{1}{\tau_{th}} + \frac{1}{\tau_b}$$

where τ_b represents the effect of the black-body radiation. The evaluation of τ_b [11] requires the knowledge of the dipole matrix elements between the level under study and all the other radiatively

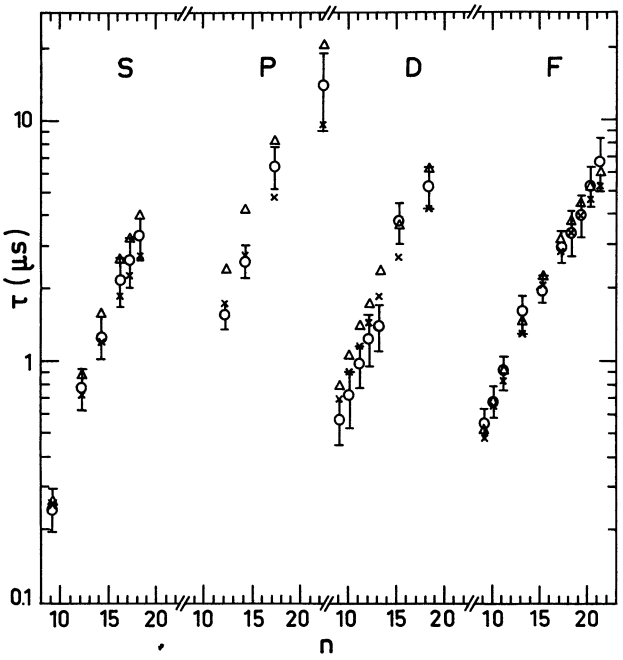

Fig. 2. — Radiative lifetimes of the rubidium S-D-F ($T = 520$ K) and P ($T = 460$ K) levels τ_{exp} : ○ ; τ_{th} : △ ; τ_{cal} (see text) : × .

connected levels. Using the B. D. method [7] for this evaluation leads to τ_{cal} values which are certainly less reliable than the τ_{th} values because the τ_b values are mainly determined by radial matrix elements connecting closely spaced Rydberg levels. It is well-known that the B.D. method is not well suited for the calculation of such matrix elements [12] which, on the other hand, contribute only a few percent in the τ_{th} values. Despite the resulting uncertainty on the τ_{cal} values it seems more appropriate for our purpose to compare the τ_{exp} values with the τ_{cal} values rather than with the corresponding τ_{th} ones. This is done in figure 2 where the τ_{cal} values are observed to exhibit close agreement with our experimental data (note that the black-body correction is small for the F levels).

For the first time a wide set of lifetime values concerning hydrogenic (i.e. F states) as well as non hydrogenic (S, P and D states) Rydberg levels is presented for a given alkali atom. Besides the interest, already mentioned, in having such data, the comparison shown on figure 2 between the τ_{exp} and τ_{cal} values supports well the conclusion that the B.D. method provides good estimates (within about 20 %) for the radiative lifetimes of the highly excited rubidium atoms, once the black-body radiation effect is taken into account. The results already obtained by other groups [1, 2, 3, 11] indicate that this conclusion probably holds also for the other alkali atoms.

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