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## Perturbation of the Ba $6sns\ ^1S_0$ sequence by the $5d7d\ ^3P_0$ state, probed by lifetime measurements

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**Abstract.** The  $6sns\ ^1S_0$  sequence of Ba is strongly perturbed around  $n = 18$  by the  $5d7d\ ^3P_0$  doubly excited state. We have probed the configuration mixing by observing the strong decrease in the measured  $^1S_0$  lifetimes around the short-lived valence perturber state. The measurements have enabled us to resolve the problem of the designation of the studied states. Wavefunctions obtained from multi-channel quantum-defect theory were used to calculate theoretical lifetimes in good agreement with the experimental values.

### 1. Introduction

In alkaline-earth atoms strong perturbations in sequences of highly excited (Rydberg) states occur due to interaction with low-lying valence states of series of the same parity, converging towards a higher series limit. Multi-channel quantum-defect theory (MQDT) (Lu and Fano 1970, Armstrong *et al* 1977) has been used to analyse recent data on energy levels (see e.g. Esherick 1977, Aymar *et al* 1978, Aymar and Robaux 1979). Configuration mixing not only affects the level positions but is also reflected in Landé factors (Wynne *et al* 1977, Grafström *et al* 1981a) and in the hyperfine structure (Beigang *et al* 1981, Grafström *et al* 1981b). Radiative properties are also very sensitive to the type of perturbations discussed here because of the great difference in lifetime between the Rydberg states (long lived) and the low-lying valence perturber states (short lived). Recently, very drastic changes in the measured lifetime values were observed for the perturbed  $6snd\ ^1,3D$  sequences of Ba (Bhatia *et al* 1981, Gallagher *et al* 1981, Aymar *et al* 1981). In the present paper similar measurements for the  $6sns\ ^1S_0$  sequence of barium are presented. The members  $n = 11-21$  were investigated together with the perturber state close to  $n = 18$ , the  $5d7d\ ^3P_0$  doubly excited state. The new lifetime measurements shed light on some problems concerning level designations (Rubbmark *et al* 1977, Aymar *et al* 1978). The wavefunctions obtained by Aymar and Robaux (1979) using MQDT were used to calculate lifetimes for the studied states which are in good agreement with the experimental values.

### 2. Experimental techniques and measurements

Our lifetime measurements were performed using the PUMOLS technique (PULSE MOdulated Laser Spectroscopy) which incorporates pulse modulation of a cw dye

laser beam and delayed-coincidence electronics (Gustavsson *et al* 1979). The experimental set-up is shown in figure 1. The even parity  $J = 0$  states were populated by stepwise laser excitation via the short-lived  $6s6p\ ^1P_1$  state employing a cw dye laser operating at  $5535\ \text{\AA}$  and a further cw dye laser operating in the wavelength region  $420\text{--}440\ \text{nm}$ . Compared with the  $^1D_2$  sequence previously investigated by Bhatia *et al* (1981) the transition probabilities to the S states are much lower than those pertinent to the  $^1D_2$  levels, necessitating the use of a single-mode dye laser for efficient excitation. The green laser was adjusted to the  $5535\ \text{\AA}$  Ba transition using optogalvanic signals from a Ba hollow cathode, whereas the single-mode dye laser was accurately set at the correct frequency employing a digital wavemeter with seven significant digits (Hertz and Nilsson 1981). Fluorescence photons were observed in the decay back to the  $6s6p\ ^1P_1$  level. An interference filter was used to select the transition. The blue dye laser was acousto-optically modulated and start/stop signals for a time-to-amplitude converter (TAC) were obtained from a photomultiplier detecting a fraction of the exciting beam and a further multiplier observing fluorescence photons from a Ba atomic beam. Pulses from the TAC were fed to a multichannel analyser, where the exponential decay curve was gradually obtained. The data were outputted on a paper punch and fitted to an exponential at an external computer.

In figure 2 an experimental curve is shown for the  $6s14s\ ^1S_0$  state together with a fitted exponential. The necessary precautions in order to avoid influences due to

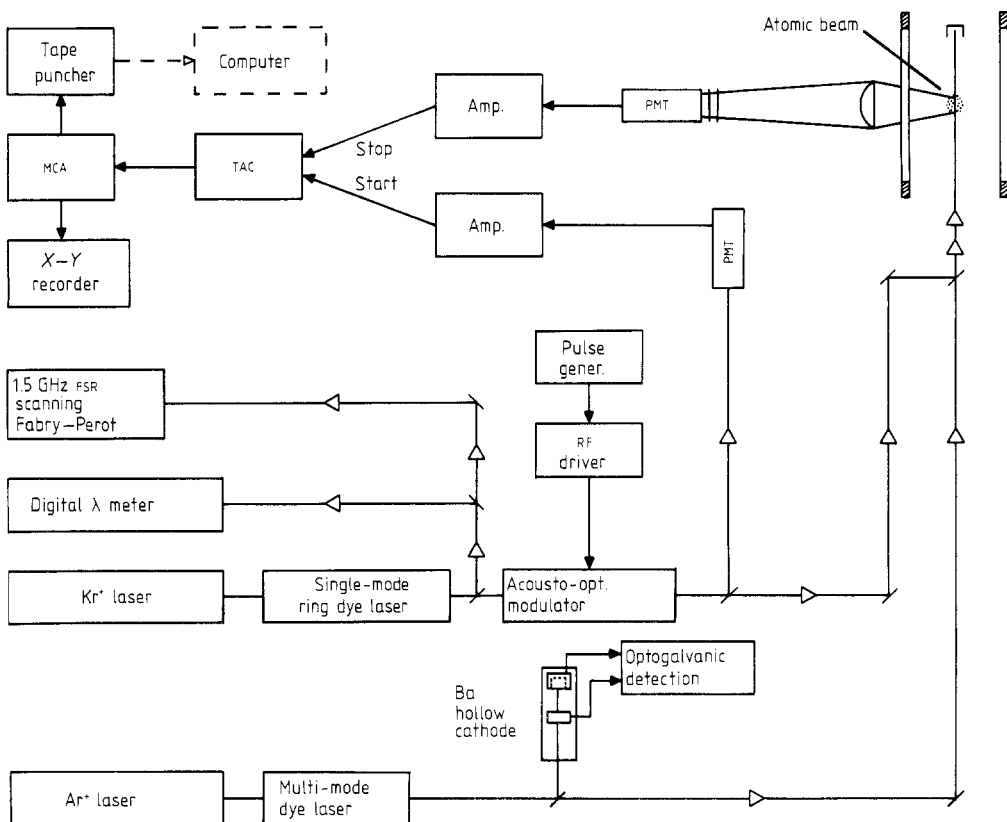
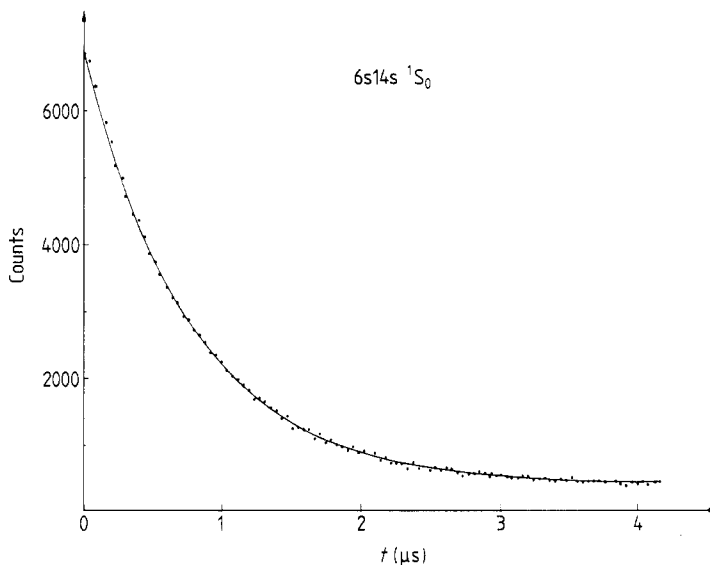


Figure 1. Experimental set-up used in the lifetime measurement of barium  $J = 0$  states.



**Figure 2.** Experimental decay curve for the  $6s14s\ ^1S_0$  state of barium.

pile-up, multiple scattering, collisions and flight-out-of-view effects were taken in the lifetime measurements (Gustavsson *et al* 1979).

The lifetime values were measured with the atomic beam exposed to the chamber walls at about 300 K. The far-infrared reflective properties of the vacuum chamber were such that the influence of the exposure to the hot oven orifice was completely negligible. The highly excited, long-lived  $^1S_0$  states will be influenced by black-body induced transitions resulting in virtually shortened natural lifetimes. The effect was found to be about 4% for  $^1D_2$  states with  $n$  around 20 (Bhatia *et al* 1981). The experimentally found value for the  $^1D_2$  states closely agrees with the corresponding theoretical results for  $^2D$  states of caesium (Farley and Wing 1981). Their calculated correction for  $^2S$  states is 2% for  $n = 10$  and 15% for  $n = 20$ . These results indicate the size of the expected correction for the studied Ba  $^1S_0$  states. The experimental, room-temperature lifetime values for  $^1S_0$  levels are given in table 1. The error bars comprise the statistical spread in the data as well as a 1.5% additional allowance for systematic errors. A strong decrease in lifetime values is very evident around  $n = 18$ .

### 3. Calculations and discussion

Some problems concerning level designations appear near the perturbation of the  $6sns\ ^1S_0$  sequence close to the  $18^1S_0$  state. Between the  $17^1S_0$  and  $19^1S_0$  levels, Rubbmark *et al* (1977) assigned two levels to the  $J = 0$  bound spectrum; a level at  $41451\text{ cm}^{-1}$  was designated as  $18^1S_0$  and a level at  $41468\text{ cm}^{-1}$  as  $5d7d\ ^1S_0$ . Aymar *et al* (1978) could not detect the former level and instead designated the level at  $41468\text{ cm}^{-1}$  as the  $18^1S_0$  state; moreover they observed a new level at  $41441\text{ cm}^{-1}$  and designated this level as the  $5d7d\ ^3P_0$  perturber. In a recent investigation (Camus *et al* 1981) two-step optical galvanic spectroscopy was used to observe the  $6sns\ ^3S_1$  series of barium and the  $41451\text{ cm}^{-1}$  level was assigned as the  $6s18s\ ^3S_1$  state. In order to

**Table 1.** Experimentally determined lifetimes (300 K) for  $J = 0$  barium states.

	State	Lifetime (ns) at 300 K
$^1S_0$ states	6s11s $^1S_0$	319 (7)
	6s12s $^1S_0$	444 (9)
	6s13s $^1S_0$	584 (13)
	6s14s $^1S_0$	753 (15)
	6s15s $^1S_0$	936 (19)
	6s16s $^1S_0$	1076 (22)
	6s17s $^1S_0$	917 (19)
	6s18s $^1S_0$	274 (20)
	6s19s $^1S_0$	2060 (60)
	6s20s $^1S_0$	3000 (200)
	6s21s $^1S_0$	3400 (300)
Perturber	5d7d $^3P_0$	138 (5)
$^3S_1$ state	6s18s $^3S_1$	2010 (50)

clarify the level designation situation we have, in addition to the lifetime measurements for  $^1S_0$  states (which included the  $41468\text{ cm}^{-1}$  state for which we have used the designation 6s18s  $^1S_0$ ), determined the lifetimes of the  $41441\text{ cm}^{-1}$  and  $41451\text{ cm}^{-1}$  states. The results are also included in table 1. Our lifetime measurements confirm the designations given by Aymar *et al* (1978) and Camus *et al* (1981). The short lifetime 138(5) ns obtained for the  $41441\text{ cm}^{-1}$  state is characteristic for a valence perturber state (5d7d  $^3P_0$ ). The lifetime value 2010(50) ns obtained for the  $41451\text{ cm}^{-1}$  state corresponds to a long-lived pure Rydberg level (6s18s  $^3S_1$ ) rather than to a short-lived perturbed state. In connection with measurements of  $g_r$  factors of barium states (Grafström *et al* 1981a) we also determined the  $g_r$  factor of this level to be close to two as expected for a  $^3S_1$  state.

The experimental lifetimes obtained for the  $J = 0$  levels have been successfully interpreted using the results provided by a MQDT analysis of the even-parity bound spectrum of Ba I (Aymar *et al* 1978). The parametric method used for interpreting experimental data is similar to that previously used by one of us (Aymar *et al* 1981) for analysing lifetime data of Rydberg levels in the perturbed 6snd  $^{1,3}D_2$  series of Ba I. The wavefunction of a  $J = 0$  bound level pertaining either to the 6sns  $^1S_0$  series or to 5dnd ( $n = 6, 7$ ) configurations can be expressed as

$$\Psi_i(\nu_i) = a_i\phi_{6ss\ ^1S_0}(\nu_i^1) + b_i\phi_{5dd\ ^1S_0}(\nu_i^2, \nu_i^3) + c_i\phi_{5dd\ ^3P_0}(\nu_i^2, \nu_i^3) \quad (1)$$

where  $a_i$ ,  $b_i$  and  $c_i$  are MQDT mixing coefficients of the 6ss  $^1S_0$ , 5dd  $^1S_0$  and 5dd  $^3P_0$  channels and the  $\phi$  functions have a pure  $LS$ -coupled angular part and a radial part involving the effective quantum number  $\nu_i$  related either to the first Ba<sup>+</sup> 6s limit ( $\nu_i^1 = n_i^*$ ) or to the Ba<sup>+</sup> 5d<sub>3/2, 5/2</sub> higher limits ( $\nu_i^2, \nu_i^3$ ). If we leave out the lowest 6sns  $^1S_0$  levels ( $11 \leq n \leq 13$ ), slightly perturbed by lower 5d6d  $J = 0$  levels, the  $b_i$  and  $c_i$  coefficients have high values only for the 5d7d  $^3P_0$  level and for some levels close to this perturber, for  $n > 13$  we can neglect the variation of  $\Psi_i$  with  $\nu_i^2$  and  $\nu_i^3$  and write

$$\Psi_i(n_i^*) = a_i\phi_{6ss\ ^1S_0}(n_i^*) + b_i\bar{\phi}_{5d7d\ ^1S_0} + c_i\bar{\phi}_{5d7d\ ^3P_0} \quad (2)$$

where the  $\bar{\phi}$  functions do not depend on  $i$ . The radiative decay rate of a given level  $i$  can be calculated from transition probabilities to lower 6snp, 5dnp and 5dnf  $J = 1$

levels. With simplifying assumptions discussed by Aymar *et al* (1981), we obtain the following expression for the radiative decay rate  $\Gamma_i$  of the level  $i$

$$\Gamma_i = a_i^2 \Gamma_{6ss\ ^1S_0}(n_i^*) + (b_i^2 + c_i^2) \Gamma_{5d7d\ ^3P_0} \tag{3}$$

where  $\Gamma_{6ss\ ^1S_0}$  is the decay rate of a level pertaining to the pure 6ss <sup>1</sup>S<sub>0</sub> channel and  $\Gamma_{5d7d\ ^3P_0}$  is the decay rate of a pure 5d7d <sup>3</sup>P<sub>0</sub> perturber, i.e. involving no admixture of the 6ss <sup>1</sup>S<sub>0</sub> channel.

Since the decay rates of pure Rydberg levels are expected to have a  $(n_i^*)^{-3}$  dependence we can write for  $i$  levels with  $n > 13$

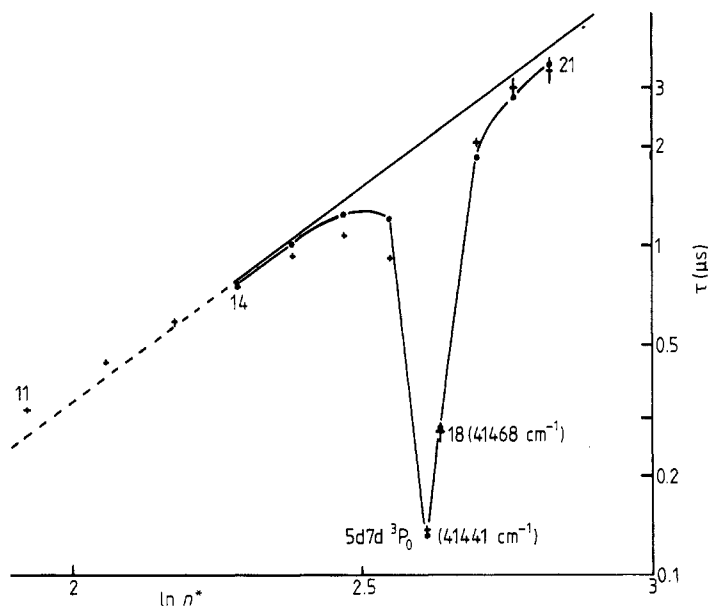
$$\Gamma_i = a_i^2 \frac{\gamma}{(n_i^*)^3} + (b_i^2 + c_i^2) \Gamma_{5d7d\ ^3P_0} \tag{4}$$

where the quantities  $\gamma$  and  $\Gamma_{5d7d\ ^3P_0}$  do not depend upon  $i$ .

For the lower levels ( $11 \leq n \leq 13$ ) we neglect the small perturbation mainly due to 5d6p perturbers and the corresponding decay rates are given by

$$\Gamma_i = \frac{\gamma}{(n_i^*)^3} \tag{5}$$

The quantities  $\gamma$  and  $\Gamma_{5d7d}$  have been determined by fitting the theoretical decay rates (equation (4) or (5)) to the experimental data. The values so obtained are  $\gamma = 1.19 \times 10^9 \text{ s}^{-1}$  and  $\Gamma_{5d7d\ ^3P_0} = 0.0113 \times 10^9 \text{ s}^{-1}$ . The comparison between theoretical and experimental lifetimes is shown in figure 3, where the results are plotted on a ln-ln diagram against the effective quantum number  $n^*$ . The straight line corresponds to equation (5). The overall agreement between experiment and theory is rather good; the drastic decrease in lifetimes around the 5d7d <sup>3</sup>P<sub>0</sub> perturber is very well



**Figure 3.** Ln-ln plot of experimental and theoretical lifetime values for  $J = 0$  states against the effective quantum number  $n^*$ . The straight line corresponds to  $\tau_i = (n_i^*)^3 / \gamma$  with  $\gamma = 1190$ . +: experimental values with error bars; ●: theoretical values.

reproduced. The new lifetime measurements allow us to confirm the designations given by Aymar *et al* (1978). For lower  $6sns\ ^1S_0$  levels ( $n \leq 13$ ) our theoretical treatment is rather rough due to the presence of the  $5d6d\ J = 0$  perturber; however, experimental data do not deviate much from the straight line describing the behaviour of unperturbed  $6sns\ ^1S_0$  levels. (Let us note that the lower levels are introduced in the fit in order to increase the number of data points from which  $\gamma$  is determined.) The fitted  $\gamma$  parameter is in good agreement with that computed by means of a central potential according to the Klapisch method (Aymar *et al* 1970):  $\gamma_{th} = 1.2 \times 10^9\ s^{-1}$ . The  $\Gamma_{5d7d\ ^3P_0}$  parameter is also in agreement with the  $\Gamma_{5d7d\ ^1D_2}$  parameter previously determined (Aymar *et al* 1981); in fact from  $\Gamma_{5d7d\ ^1D_2} = 0.015 \times 10^9\ s^{-1}$  one can deduce  $\Gamma_{5d7d\ ^3P_0} = 0.01 \times 10^9\ s^{-1}$  taking into account a wavelength factor leading to a smaller decay rate for the lower  $5d7d\ ^3P_0$  level.

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