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Perturbation of the Ba 6s*n*s $^1S_0$ sequence by the 5d7d $^3P_0$ state, probed by lifetime measurements

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Abstract. The 6s*n*s $^1S_0$ 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 6s*n*d $^{1,3}D$ sequences of Ba (Bhatia *et al* 1981, Gallagher *et al* 1981, Aymar *et al* 1981). In the present paper similar measurements for the 6s*n*s $^1S_0$ 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

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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 Å and a further CW dye laser operating in the wavelength region 420–440 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 Å 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

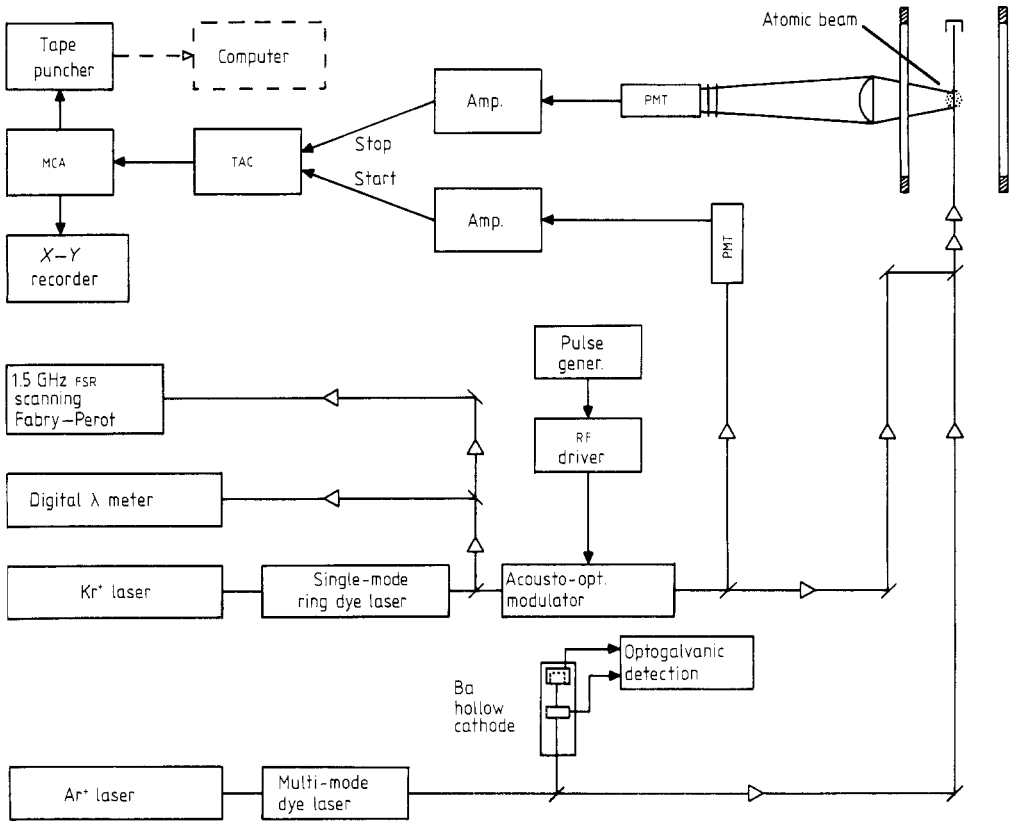

Figure 1. Experimental set-up used in the lifetime measurement of barium $J = 0$ states.

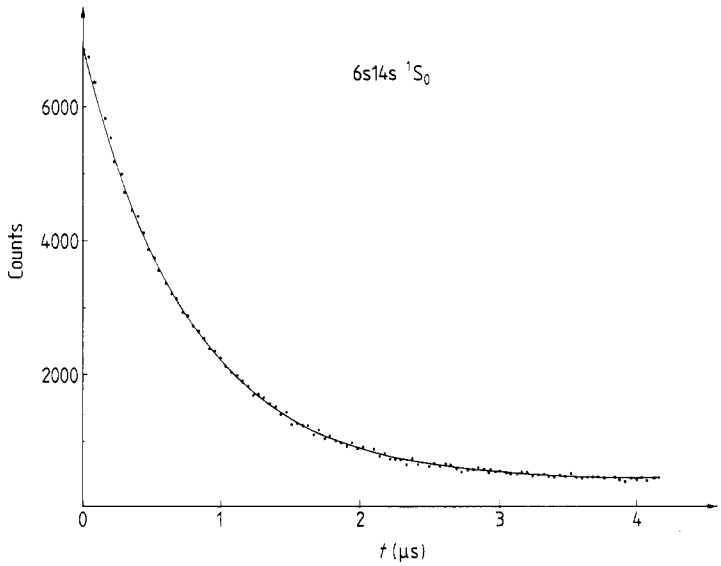

Figure 2. Experimental decay curve for the 6s14s $^1S_0$ state of barium.

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 cm^{-1} was designated as 18^1S_0 and a level at 41468 cm^{-1} as 5d7d $^1S_0$. Aymar *et al* (1978) could not detect the former level and instead designated the level at 41468 cm^{-1} as the 18^1S_0 state; moreover they observed a new level at 41441 cm^{-1} and designated this level as the 5d7d $^3P_0$ perturber. In a recent investigation (Camus *et al* 1981) two-step optogalvanic spectroscopy was used to observe the 6sns $^3S_1$ series of barium and the 41451 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 cm^{-1} state for which we have used the designation 6s18s $^1S_0$), determined the lifetimes of the 41441 cm^{-1} and 41451 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 cm^{-1} state is characteristic for a valence perturber state (5d7d $^3P_0$). The lifetime value 2010(50) ns obtained for the 41451 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_J factors of barium states (Grafström *et al* 1981a) we also determined the g_J 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

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

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 ϕ functions have a pure LS-coupled angular part and a radial part involving the effective quantum number ν_i related either to the first Ba^+ 6s limit ($\nu_i^1 = n_i^*$) or to the Ba^+ $5d_{3/2,5/2}$ higher limits (ν_i^2, ν_i^3). If we leave out the lowest 6sns $^1S_0$ levels ($11 \leqslant n \leqslant 13$), 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 Ψ_i with ν_i^2 and ν_i^3 and write

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

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 Γ_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 $^1S_0$ channel and $\Gamma_{5d7d\,^3P_0}$ is the decay rate of a pure 5d7d $^3P_0$ perturber, i.e. involving no admixture of the 6ss $^1S_0$ 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 γ and $\Gamma_{5d7d\,^3P_0}$ do not depend upon i.

For the lower levels ($11 \leqslant n \leqslant 13$) 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 γ and Γ_{5d7d} have been determined by fitting the theoretical decay rates (equation (4) or (5)) to the experimental data. The values so obtained are $\gamma = 1.19 \times 10^9\ s^{-1}$ and $\Gamma_{5d7d\,^3P_0} = 0.0113 \times 10^9\ s^{-1}$. 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 $^3P_0$ perturber is very well

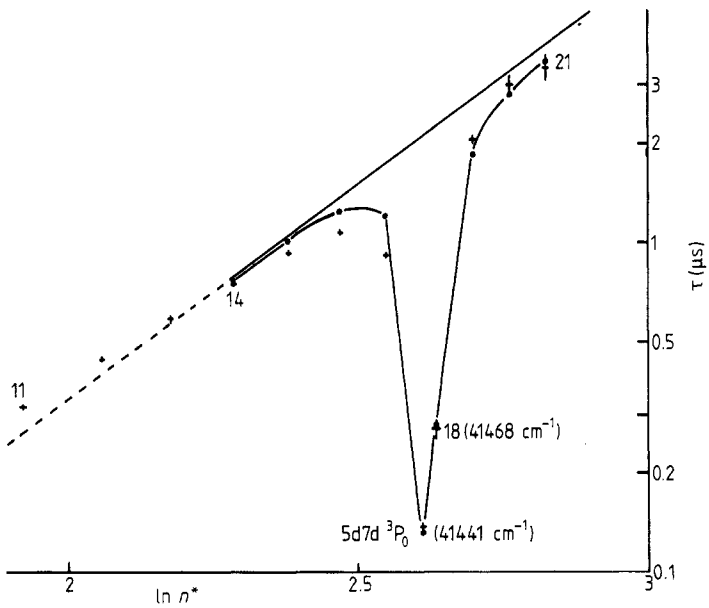

Figure 3. Ln–ln plot of experimental and theoretical lifetime values for $J = 0$ states against the effective quantum number n^*. The straight line corresponds to $\tau_i = (n_i^*)^3/\gamma$ with $\gamma = 1190$. +: experimental values with error bars; ●: theoretical values.

reproduced. The new lifetime measurements allow us to confirm the designations given by Aymar *et al* (1978). For lower $6sns\ ^1S_0$ levels ($n \leqslant 13$) 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 γ is determined.) The fitted γ 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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