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DETERMINATION OF NATURAL RADIATIVE LIFETIMES FOR S AND D STATES IN RUBIDIUM AND CESIUM USING A PULSED DYE LASER

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Using delayed-coincidence techniques we have measured the natural radiative lifetimes of several S and D states in Rb and Cs. The states were excited by a pulsed dye laser from the first P state, populated using an rf discharge lamp.

Through the availability of tunable lasers, spectroscopic investigations of atomic states have been greatly facilitated. Using step-wise excitation in combination with lasers, states normally inaccessible with conventional light sources can be studied. The alkali atoms are particularly interesting as they have a simple structure suitable for theoretical calculations. A usual approach is to treat the alkali atom as a hydrogen-like system, with an external electron moving in a central field produced by the nucleus and closed inner shells. Whereas such a model quite well explains the hyperfine structure of S and P states it completely fails describing the hyperfine structure of D states, which has been found to be highly anomalous [e.g. 1, 2]. Detailed calculations of e.g. many-body-perturbation type are necessary to explain the observed structures. On the other hand, properties like radiative lifetimes and Stark effect parameters depending on transition matrix elements $\langle \Psi | r | \phi \rangle$ are likely to be well described by hydrogen-like wavefunctions. This is due to the fact that the properties of the wavefunctions far off the nucleus are the most important in these calculations, contrary to what regards the case of hyperfine structure. It is interesting to find out in experiments to what extent the hydrogen-like model is adequate.

In the present experiments the lifetimes of several S and D states in rubidium and cesium were determined. The states were reached using step-wise excitation, utilizing an rf lamp to populate the first $^2P$ level and a dye laser for the subsequent excitation. This technique was used in several double-resonance and level-crossing measurements at Columbia University and in this laboratory [e.g. 3, 4]. This excitation scheme has recently been used by Deech et al. in quantum-beat studies of the 8, 9, and $^{10}2D_{3/2}$ states in cesium [5]. Apart from hyperfine coupling constants, accurate lifetime values were derived in delayed-coincidence measurements. Further, for sodium, the lifetime of a large number of S and D states have recently been measured in step-wise excitation experiments involving two pulsed dye lasers [6, 7].

In fig. 1 our experimental set-up is schematically shown. It is similar to the one used by Deech et al. A sealed-off resonance cell, containing $^{87}$Rb or $^{133}$Cs, was heated in an oven by a stream of hot air. A powerful rf lamp was used to populate the first P state. A Molelectron DL 400 dye laser, pumped by a Moletron UV 400 N$_2$ laser was utilized for the subsequent excitation to an S or D state. The dye laser had a linewidth of about 0.1 Å. The repetition rate was usually about 50 Hz, and the pulse-length 5 ns. Fluorescent photons, released after the pulse excitation, were selected for wavelength by a grating monochromator and detected by a Peltier-cooled EMI 9558 BQ photomultiplier tube. A trigger signal for the electronics was derived from a diode, detecting the laser pulse. A PAR 162 boxcar integrator with a time resolution of 10 ns was used to monitor the exponential decay down to the first excited P state. Alternatively, a time-to-pulse-height converter (TAC) was used in delayed-coincidence measurements. The start signal was obtained from the diode, and the stop signal from the first fluorescent photon, detected by the photomultiplier. In these experiments neutral filters in the laser beam were used to ensure that the probability for detecting a photon, following a single laser shot was 10 per cent or less. Thus the pile-up correction could be accurately accounted for [8]. The output pulses
from the TAC were stored in a multichannel analyser. The accumulated signal was corrected for pile-up and fitted to an exponential using a computer program. In fig. 2 an experimental curve is shown for the $^9 2S_{1/2}$ state of Rb. The curves from the boxcar integrator were analyzed manually. Series of measurements using either detection systems were made for the studied states. Great care was taken to ensure that the cell was operated in a temperature range, where collision effects were negligible. Ordinary working temperatures were in the region 70–80°C. Radiation trapping was absent, as the terminal state is a weakly populated excited state. Polarizers were used in the laser beam and detection beam to eliminate hyperfine beats as discussed by Deech et al. [5]. For each state the experiments were performed in approximately zero magnetic field as well as at 10 Gauss. No changes in the curve shape were observed and thus Zeeman beats did not influence our measurements.

In table 1 we give our results for the measured states. Our values are mainly based on the delayed-coincidence experiments which we found to be the most reliable. We compare our results with theoretical values, calculated using hydrogen-like wavefunctions [9]. It is found that substantial deviations between experiment and theory exist, which on the other hand is not observed in the Stark interaction [2]. More detailed

![Diagram of experimental set-up](https://example.com/diagram)

**Fig. 1.** Experimental set-up used in the lifetime measurements.

![Graph of experimental decay curve](https://example.com/graph)

**Fig. 2.** Experimental decay curve obtained for the $^9 2S_{1/2}$ state in Rb.

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<tbody>
<tr>
<td>Rb</td>
<td>$^8 2S_{1/2}$</td>
<td>153(8)</td>
<td>188</td>
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<tr>
<td></td>
<td>$^9 2S_{1/2}$</td>
<td>258(13)</td>
<td>324</td>
</tr>
<tr>
<td></td>
<td>$^6 2D_{3/2}$</td>
<td>285(16)</td>
<td>215</td>
</tr>
<tr>
<td></td>
<td>$^7 2D_{3/2}$</td>
<td>388(25)</td>
<td>264</td>
</tr>
<tr>
<td></td>
<td>$^8 2D_{3/2}$</td>
<td>515(30)</td>
<td>347</td>
</tr>
<tr>
<td>Cs</td>
<td>$^{10} 2S_{1/2}$</td>
<td>260(12)</td>
<td>324</td>
</tr>
<tr>
<td></td>
<td>$^{11} 2S_{1/2}$</td>
<td>343(22)</td>
<td>514</td>
</tr>
<tr>
<td></td>
<td>$^{12} 2S_{1/2}$</td>
<td>545(30)</td>
<td>779</td>
</tr>
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</table>

In table 1 we give our results for the measured states. Our values are mainly based on the delayed-coincidence experiments which we found to be the most reliable. We compare our results with theoretical values, calculated using hydrogen-like wavefunctions [9]. It is found that substantial deviations between experiment and theory exist, which on the other hand is not observed in the Stark interaction [2]. More detailed
theoretical calculations, also including the spin-orbit perturbation [10], are needed to shed light upon these observations.

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References

W. Happer and P. Tsekeris, private communication.