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Natural radiative lifetimes in the interacting 5*snd* $^{1,3}D_2$ sequences in Sr

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Natural radiative lifetimes have been determined in the $5snd\ ^{1,3}D_2$ sequences of strontium in the perturbed region around $n = 15$. Stepwise laser excitations and pulse modulation of a cw dye-laser beam were employed. A plot of the measured lifetime values versus the effective quantum number reveals complex configuration interaction.

I. INTRODUCTION

The alkaline-earth atoms have two electrons outside closed shells and the presence of low-lying, doubly excited states results in strong perturbations of sequences with only one excited electron. These configuration-interaction effects can be studied not only in the basic energy-level structure but are also reflected in, e.g., radiative lifetimes, hyperfine structures, and isotope shifts. For a theoretical description of these effects, the multichannel quantum-defect theory (MQDT) is usually applied.

In strontium, the energy levels of the $5snd\ ^{1,3}D_2$ sequences together with perturbing states have been measured by several authors.[1,2,3] In Ref. 2 the data were analyzed with the use of MQDT. The g_J factors, calculated from the obtained wave functions, were found to agree very well with experimental values determined in a Zeeman-effect investigation in the perturbed region around $n = 15$.[4] In the same region, a strong variation in the hyperfine interaction has also been found.[5]

In the present work we have measured radiative lifetimes in the $5snd\ ^1D_2$, $n = 13$–22, and $5snd\ ^3D_2$, $n = 14$–17 sequences of strontium using the PUMOLS technique (pulse modulated laser spectroscopy).[6] Stepwise excitations of a collimated atomic beam were employed. By using a single-mode laser for the second step a selective excitation of the isotope ^{88}Sr was performed. Lifetime values for lower-energy members of the $^1D_2$ sequence and some low-lying $^1S_0$ levels have been measured by Gornik.[7]

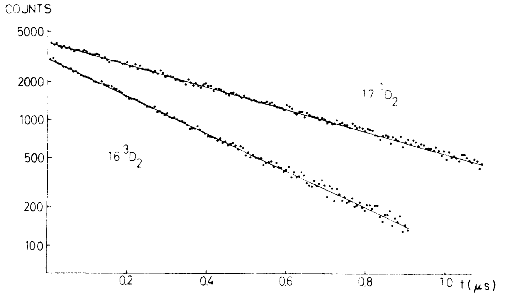

FIG. 1. Experimental decay curves for the Sr states $5s\,17d\ ^1D_2$ and $5s\,16d\ ^3D_2$. Background counts are subtracted.

For radiative lifetimes, properties of the wave functions other than those for energy levels and g_J factors are important, and it is interesting to investigate how well different manifestations of perturbation can be reproduced. In recent measurements on barium regarding lifetimes[8] and g_J factors,[9] published MQDT wave functions[10] were found to describe the radiative properties well but resulted in inadequate predictions of the g_J factors. This has resulted in a refinement in the MQDT treatment,[9,11] leading to a much improved g_J description.

II. EXPERIMENTAL ARRANGEMENT

The experimental setup used in the present measurements was similar to that described in Ref. 8. A blue multimode cw dye laser, operating with the dye Coumarin 1, induced the 4607-Å transition from the ground state $5s^2\ ^1S_0$ to the intermediate $5s5p\ ^1P_1$ level, which has a lifetime of only 4.8 ns.[12] A single-mode ring dye laser, operating with the dye Stilbene 3, was used for the second step from the $^1P_1$ to the $^{1,3}D_2$ states. Both dye lasers were pumped by ion lasers with uv lines. The output beam of the single-mode dye laser was pulse-modulated by an acousto-optic modulator for delayed-coincidence measurements. The atomic beam of low collimation came from a resistively heated oven containing Sr metal. The two laser beams were made to overlap and crossed the atomic beam at about a right angle. The decay of the investigated level was detected with a cooled photomultiplier tube equipped with suitable filters. A photon, detected by the photomultiplier, was used as a "stop" signal, whereas a signal derived from the prompt pulse was used as a "start" signal for a time-to-amplitude converter. The pulses produced by this unit were fed to a multichannel analyzer (Tracor Northern TN 1710).

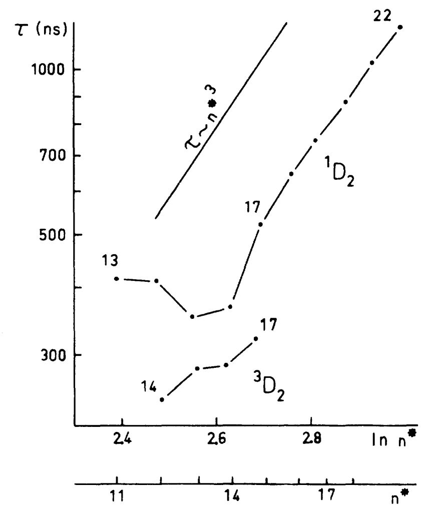

FIG. 2. Measured lifetimes in the Sr $^{1,3}D_2$ sequences plotted vs the effective principal quantum number.

III. MEASUREMENTS AND RESULTS

In the measurements the first-step laser was tuned to the 4607-Å transition from the $5s^2\,{}^1S_0$ to the $5s5p\,{}^1P_1$ state by observing the intense blue fluorescence light. The wavelengths of the second-step laser were measured with a digital wavelength meter and were found to be in good agreement with Ref. 3. For each individual lifetime value a suitable pulse length and repetition rate was set on the acousto-optic modulator in order to get an optimum duty cycle. Since the wavelengths of the first and the second step are fairly close, complete suppression of the intense blue resonance fluorescence was sometimes hard to achieve when we detected at the laser wavelength. On some occasions uv detection of the decay to the $5s5p\,{}^3P$ levels was used resulting in a more efficient suppression. Experimental decay curves for the $5s16d\,{}^3D_2$ and $5s17d\,{}^1D_2$ states are shown in Fig. 1. The multichannel analyzer was interfaced to a minicomputer and every decay curve was stored as a data file on a floppy disc. This provided us with the opportunity of calculating the radiative lifetime of one decay curve while another one was being stored in the multichannel analyzer. For each state, measurements were performed several times and particular attention was paid to ensure that there was no influence due to pileup, multiple scattering, collisions, Zeeman quantum beats, and flight-out-of-view effects.[6]

TABLE I. Experimentally determined natural radiative lifetimes for Sr states (300 K).

State	Measured τ value (ns)
$5s13d\,{}^1D_2$	410(20)
$5s14d\,{}^1D_2$	408(12)
$5s15d\,{}^1D_2$	340(10)
$5s16d\,{}^1D_2$	365(15)
$5s17d\,{}^1D_2$	517(16)
$5s18d\,{}^1D_2$	640(16)
$5s19d\,{}^1D_2$	738(37)
$5s20d\,{}^1D_2$	866(40)
$5s21d\,{}^1D_2$	1023(51)
$5s22d\,{}^1D_2$	1190(60)
$5s14d\,{}^3D_2$	247(8)
$5s15d\,{}^3D_2$	282(11)
$5s16d\,{}^3D_2$	286(9)
$5s17d\,{}^3D_2$	319(15)
$4d^2\,{}^3P_2$	7(2.5)
$5s15s\,{}^1S_0$	1145(57)
$5s16s\,{}^1S_0$	1424(72)
$5s7f\,{}^1F_3$	126(5)
$5s10p\,{}^1P_1$	92(5)

The experimentally determined natural radiative lifetimes for the Sr sequences $5snd\,{}^1D_2$ ($n=13$—22) and $5snd\,{}^2D_2$ ($n=14$—17) can be seen in Table I and Fig. 2. The states $5s15s\,{}^1S_0$, $5s16s\,{}^1S_0$, $5s7f\,{}^1F_3$, and $5s10p\,{}^1P_1$ could also be reached in the employed laser wavelength region and the corresponding lifetime values are included in Table I. The last two of these states were excited from the metastable $5s4d\,{}^1D_2$ level, populated in the cascade decay of the $5s5p\,{}^1P_1$ level. The given lifetime values are referred to room temperature and are not corrected for expected small effects of black-body radiation (see, e.g., Refs. 13 and 8). The lifetime of the doubly excited perturber state $4d^2\,{}^3P_2$ was also determined. In order to measure this particularly short lifetime we first recorded the shape of the laser pulse from the acousto-optic modulator by detecting reflected light inside the vacuum system. With uv detection, we recorded the time behavior of the fluorescent light from the state. With a computer program we could generate. from the pulse shape, decay curves for different lifetimes and make the best fit to our experimental curve.

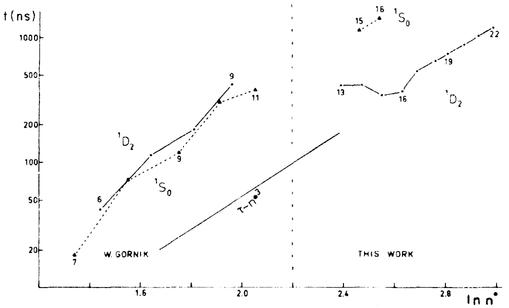

FIG. 3. Diagram including previously determined (Ref. 7) and currently measured lifetimes in the Sr $^1D_2$ and $^1S_0$ sequences.

IV. DISCUSSION

In Fig. 2 the lifetimes for the ${}^{1,3}D_2$ sequences are plotted versus the effective principal quantum number. As can be seen, there is a decrease in the lifetime values around $n=15$ for the ${}^{1}D_2$ sequence. In this energy range the $5snd$ ${}^{13}D_2$ levels are perturbed by the $4d6s$ ${}^{1,3}D_2$ levels; moreover the perturbers induce a large singlet-triplet mixing.[2] In the figure a line with the slope of three is also included. If the effect of perturbations is neglected, the lifetimes in a Rydberg sequence are expected to scale with a power, close to three, of the effective principal quantum number. However, to be in an overall accordance with this rule, the determined values for the $13\,{}^{1}D_2$ and $14\,{}^{1}D_2$ states are too long. In Fig. 3 our ${}^{1}D$- and ${}^{1}S$-state lifetime values are plotted together with the data for lower states, investigated by Gornik.[7] A slope-of-three line is also included. In this plot it is evident that complex perturbations are present. MQDT calculations[2] have shown that the $4d6s$ ${}^{1,3}D_2$ perturbation is spread out over a large number of Rydberg levels. It would be very worthwhile to see if the apparent shortening of the $6snd$ ${}^{1}D_2$ lifetime values for $n \geq 13$ with respect to Gornik's values can be explained by taking this mixing into account.

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