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Lifetime measurements in the ${}^2S_{1/2}$ and ${}^2D_{3/2,5/2}$ sequences of indium

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Natural radiative lifetimes in the $5s^2ns\ {}^2S_{1/2}$ and $5s^2nd\ {}^2D_{3/2,5/2}$ ($n \leq 20$) sequences of indium have been measured using pulsed laser excitation of an atomic beam. The uv pulses were produced by a YAG-pumped, or alternatively, an excimer-pumped dye-laser system. While the S -state lifetimes show a normal $(n^*)^3$ behavior, the D sequences are strongly perturbed. The lifetimes of the $5s\ 5p^2\ {}^4P$ states were found to be J dependent.

I. INTRODUCTION

During the last few years, highly excited atomic states have been extensively studied regarding radiative properties as well as energy substructure. Using laser-spectroscopy techniques, especially, the alkali-metal and alkaline-earth elements have been investigated. In this paper we report on lifetime measurements on the group-III element In. Indium has three electrons outside closed shells but two electrons usually form a closed subshell. The basic spectrum is then alkali-metal-like with a 2P state as ground level. The group-III atoms are well suited for laser-spectroscopic investigations since the 2S and 2D sequences are directly accessible from the ground configuration in one-step excitation procedures provided short uv laser-wavelengths can be obtained. In our measurements, lifetimes in the $5s^2ns\ {}^2S_{1/2}$ and $5s^2nd\ {}^2D_{3/2,5/2}$ sequences up to $n = 20$ were determined. Two different laser systems were used alternatively; a Nd:YAG-pumped Rhodamine dye laser employing frequency doubling and anti-Stokes Raman shifting, or an excimer-pumped Coumarine-47 dye laser employing frequency doubling.

Although alkali-metal-like, the In spectrum is complicated by the presence of states with two excited electron orbitals. The $5s\ 5p^2$ configuration has states both above and below the ionization limit and can cause perturbations in the Rydberg sequences. Such perturbations have been studied very extensively in the alkaline-earth elements and influence lifetime values¹ as well as g_J factors,² hyperfine structures, and isotope shifts.^{3,4}

The basic energy-level structure in In has been determined by Garton and Codling in absorption measurements.⁵ Recently the investigations were extended to high principal quantum numbers using laser-spectroscopy methods.⁶ The energy levels were

found to be affected by perturbations. Only a few hyperfine structures have been measured so far. The hyperfine structure in the lowest 2D states, measured by the level-crossing technique, was found influenced by interaction with the $5s\ 5p^2$ configuration,^{7,8} while the hyperfine structure in the lowest 2P states is affected by core polarization effects.⁹ Very recently the lifetimes of the lowest members of the 2S and 2D sequences have been measured.¹⁰

II. EXPERIMENTAL TECHNIQUE

The setup used in the present investigation is shown in Fig. 1. During the measurements we used one out of the two laser systems shown. The frequency-doubled green light from a Nd:YAG-laser (Quanta Ray DCR-1A) was used for pumping a dye laser (Quanta Ray PDL-1) with Rhodamine-590, Rhodamine-640, or Kiton red dye. The 532-nm pump pulses had an energy of about 300 mJ. The dye laser light was frequency doubled into the uv region using a KD*P (deuterated potassium dihydro-

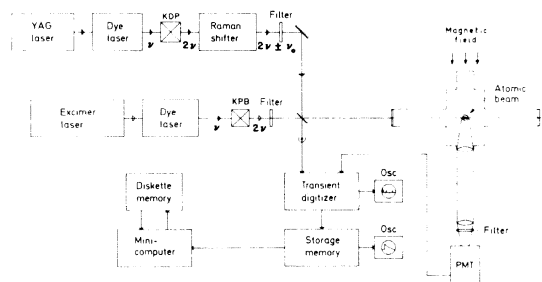


FIG. 1. Experimental setup showing the two laser systems used alternatively.

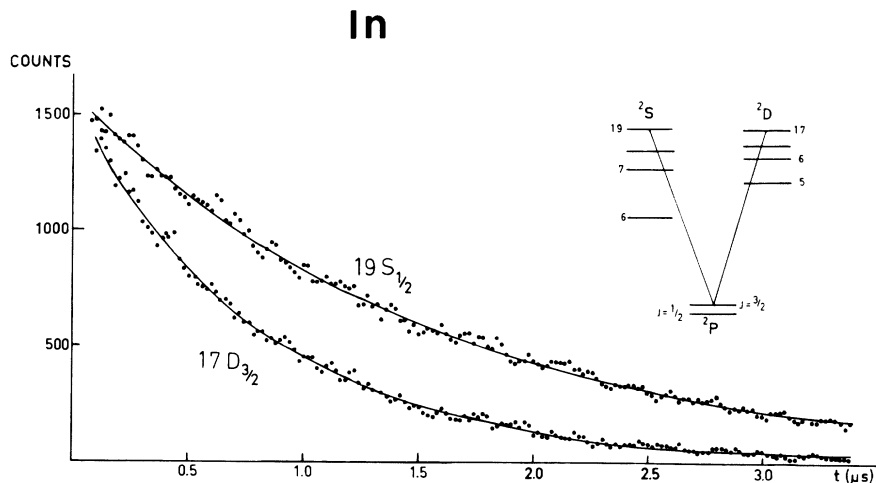


FIG. 2. Experimental data points in arbitrary units and fitted exponentials for $19^2S_{1/2}$ and $17^2D_{3/2}$. Data were sampled for 2000 laser shots. A schematic energy-level diagram is included.

gen phosphate) crystal. The uv light was passed through a Raman-shifter (Quanta Ray RS-1) operating with 10 atm of H_2 gas. Every Raman shift changes the photon energy by 4155 cm^{-1} . Most of the light passes through unshifted, but the Stokes components decrease energy and the anti-Stokes components increase energy. With a suitable laser dye, frequency doubling, and combination with up to second-order anti-Stokes shifting, we could produce tunable light for all the studied transitions 227–451 nm. The excitation energy varied between 0.01 and 20 mJ depending on the laser dye and the nonlinear process. In some cases we used a mixture between Rhodamine-610 and Rhodamine-590 dyes to optimize the power in critical wavelength ranges. The major part of the measurements were made with the Nd:YAG-laser system and all the lifetimes were measured at least at two different occasions using this system.

In some of the measurements we used an excimer laser (Lambda Physik EMG 102) with XeCl gas mixture pumping a dye laser (Lambda Physik FL 2002) with Coumarine-47 dye. The dye laser light was frequency doubled with a KPB ($KB_5O_8 \cdot 4H_2O$) crystal. The 308-nm pump pulses had an energy of about 150 mJ using 10 Hz repetition rate. The tunable uv pulses had an energy of approximately 0.01 mJ at 230 nm. The smaller pulse energy generated by the excimer laser system was to some extent compensated by a narrower spectral width.

An atomic beam of In was created in a high-vacuum system. The indium atoms were excited from the $^2P_{1/2}$ ground or the $^2P_{3/2}$ thermally populated metastable state to the $^2S_{1/2}$ and $^2D_{3/2,5/2}$ states. The exciting light was passed in and out of the vacuum system through long tubes in order to reduce stray light. The fluorescent light from the excited states was analyzed using narrow interfer-

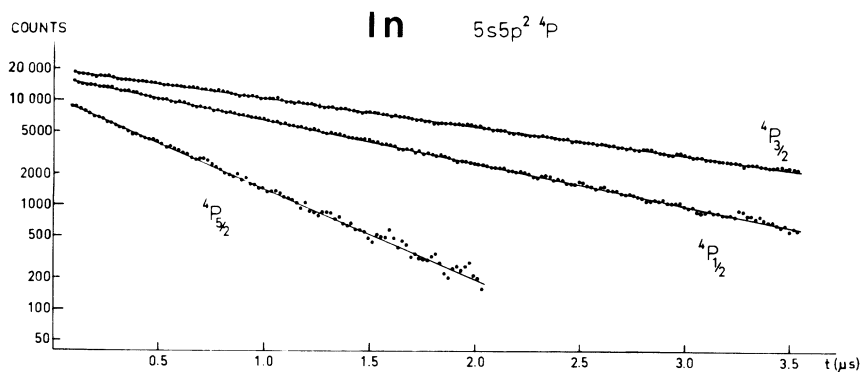


FIG. 3. Logarithms of the decay curves for the three states of the $5s5p^2 4P$ term.

TABLE I. Experimental values for natural radiative lifetimes. Excitation wavelengths and the methods of wavelength generation are included. Normally the excitation starts from the $^2P_{3/2}$ metastable level.

State	Lifetime (ns)		Wavelength (Å)	Specifics of Nd:Yag-pumped dye-laser excitation	Additional measurements with excimer system (X)
	this work	other works			
$5s^2 6s^2 S_{1/2}$		7.0(3) ^b	4511.31		
$5s^2 7s^2 S_{1/2}$	27(6)	19.5(15) ^c	2753.88 ^a	R590,D	
$5s^2 8s^2 S_{1/2}$	55(6)	53(5) ^c	2601.75	KR,D,1AS	
$5s^2 9s^2 S_{1/2}$	104(12)	118(10) ^c	2468.02	R590,D,1AS	X
$5s^2 10s^2 S_{1/2}$	163(13)	200(20) ^c	2278.20 ^a	R590,D,2AS	
$5s^2 11s^2 S_{1/2}$	244(19)		2358.70	KR,D,2AS	
$5s^2 12s^2 S_{1/2}$	330(21)		2332.76	KR,D,2AS	X
$5s^2 13s^2 S_{1/2}$	490(35)		2315.09	R590 + R610,D,2AS	X
$5s^2 14s^2 S_{1/2}$	625(60)		2302.49	R590,D,2AS	X
$5s^2 15s^2 S_{1/2}$	785(70)		2293.17	R590,D,2AS	X
$5s^2 16s^2 S_{1/2}$	1025(70)		2286.09	R590,D,2AS	X
$5s^2 17s^2 S_{1/2}$	1170(95)		2280.59	R590,D,2AS	
$5s^2 18s^2 S_{1/2}$	1360(135)		2276.21	R590,D,2AS	
$5s^2 19s^2 S_{1/2}$	1690(200)		2272.67	R590,D,2AS	
$5s^2 20s^2 S_{1/2}$	2000(300)		2269.80	R590,D,2AS	
$5s^2 5d^2 D_{3/2}$		7.0(4) ^d	3258.56		
$5s^2 6d^2 D_{3/2}$		25.5(10) ^d	2713.94		
$5s^2 7d^2 D_{3/2}$	200(14)		2522.98	R590,D,1AS	
$5s^2 8d^2 D_{3/2}$	317(22)		2430.99	R640,D,2AS	X
$5s^2 9d^2 D_{3/2}$	550(50)		2379.00	KR,D,2AS	X
$5s^2 10d^2 D_{3/2}$	455(40)		2346.56	KR,D,2AS	X
$5s^2 11d^2 D_{3/2}$	490(50)		2324.92	KR,D,2AS	X
$5s^2 12d^2 D_{3/2}$	485(40)		2309.75	R590,D,2AS	X
$5s^2 13d^2 D_{3/2}$	500(30)		2298.70	R590,D,2AS	
$5s^2 14d^2 D_{3/2}$	570(40)		2290.42	R590,D,2AS	
$5s^2 15d^2 D_{3/2}$	635(40)		2283.98	R590,D,2AS	
$5s^2 16d^2 D_{3/2}$	735(60)		2278.96	R590,D,2AS	
$5s^2 17d^2 D_{3/2}$	820(65)		2274.94	R590,D,2AS	
$5s^2 18d^2 D_{3/2}$	895(60)		2271.65	R590,D,2AS	
$5s^2 19d^2 D_{3/2}$	1075(70)		2268.94	R590,D,2AS	
$5s^2 20d^2 D_{3/2}$	1275(115)		2266.70	R590,D,2AS	

ence filters and detected with a photomultiplier tube (EMI 9558 BQ). Both excitation and detection were at right angles to the atomic beam.

After every laser pulse the fluorescence decay was captured by a transient digitizer (Biomation 8100). The transient digitizer was triggered by a photodiode detecting a small fraction of the dye-laser light. Every channel in the transient digitizer corresponds to 10 ns. The time scale was calibrated using a radio frequency signal. The digitized data was transferred to a storage memory where a large number of transients could be added. In order to get a good signal-to-noise ratio about 1000 transients were added before the signal was transferred to a mini-computer (Digital Equipment Corporation PDP-11V03) and stored on a floppy disc.

III. MEASUREMENTS AND RESULTS

The excitation wavelengths for reaching the individual states were calculated from the data given in Refs. 11 and 5. The actual wavelength setting for the dye laser was calculated considering the frequency doubling and Raman shifting. To avoid the influence of slow Zeeman quantum beats, a set of Helmholtz coils supplied a sufficient magnetic field in the excitation volume to cause beats far beyond the high-frequency out-off of the detection system. To reduce radio frequency disturbances originating from the pump laser (especially the excimer laser) we used a very short cable between the photomultiplier tube and the *A* input of the transient digitizer. With a pickup cable of suitable length in the *B* input

TABLE I. (Continued.)

State	Lifetime (ns)		Wavelength (Å)	Specifics of Nd:Yag-pumped dye-laser excitation	Additional measurements with excimer system (X)
	this work	other works			
$5s^25d^2D_{5/2}$		7.1(6) ^c	3256.09		
$5s^26d^2D_{5/2}$		18.6(15) ^c	2710.27		
$5s^27d^2D_{5/2}$	147(10)	154(10) ^c	2521.37	R590,D,1AS	
$5s^28d^2D_{5/2}$	238(20)	300(60) ^c	2429.86	R640,D,2AS	X
$5s^29d^2D_{5/2}$	640(50)		2378.14	KR,D,2AS	X
$5s^210d^2D_{5/2}$	780(80)		2345.90	KR,D,2AS	X
$5s^211d^2D_{5/2}$	760(80)		2324.41	KR,D,2AS	X
$5s^212d^2D_{5/2}$	780(65)		2309.32	R590,D,2AS	X
$5s^213d^2D_{5/2}$	845(60)		2298.33	R590,D,2AS	X
$5s^214d^2D_{5/2}$	1005(70)		2290.10	R590,D,2AS	X
$5s^215d^2D_{5/2}$	1185(110)		2283.75	R590,D,2AS	X
$5s^216d^2D_{5/2}$	1335(125)		2278.80	R590,D,2AS	X
$5s^217d^2D_{5/2}$	1540(200)		2274.79	R590,D,2AS	
$5s^218d^2D_{5/2}$	1810(210)		2271.55	R590,D,2AS	X
$5s^219d^2D_{5/2}$	1985(250)		2268.87	R590,D,2AS	
$5s^220d^2D_{5/2}$	2215(250)		2266.65	R590,D,2AS	
$5s5p^2^4P_{1/2}$	1065(65)		2858.13 ^a	R590,D	
$5s5p^2^4P_{3/2}$	1645(110)		2775.36 ^a	R590,D	
$5s5p^2^4P_{5/2}$	510(30)		2836.70	R590,D	

^aExcitation from the ground level ${}^2P_{1/2}$; R, Rhodamine; KR, Kiton Red; D, frequency doubling; 1AS, 1 anti-Stokes shift; 2AS, 2 anti-Stokes shifts.

^bReference 14.

^cReference 10.

^dReference 7.

^eReference 8.

of the transient digitizer we could eliminate the radio frequency disturbances using an A -minus- B option.

When a decay curve had been transferred to the minicomputer, a program was used for obtaining the best exponential fit to the experimental data. On some short-lived states we recorded both the fluorescent light and the exciting laser pulse. From the laser pulse shape, a computer program generated decay curves for different lifetimes to make the best fit to our experimental decay curve. Figures 2 and 3 show the quality of some experimental curves. Background counts are subtracted and the fitted exponentials are included.

For each state, recordings were made on several occasions and some states were measured with both laser systems. Special attention was paid to ensure that there was no influence due to saturation in the photomultiplier tube, multiple scattering, collisions, Zeeman quantum beats, and flight-out-of-view effects.¹² Table I gives our experimentally determined lifetimes for the ${}^2S_{1/2}$ and ${}^2D_{3/2,5/2}$ Rydberg sequences and the doubly excited configuration

$5s5p^2^4P$. The table includes excitation wavelengths, wavelength-generating system, and lifetime values measured by other groups. The error bars in our measurements include both statistical scattering and estimated possible systematic error effects in the used method. No correction has been applied for transitions induced by blackbody radiation (see, e.g., Ref. 13). Thus the given values pertain to room temperature. The blackbody correction is probably of the order of a few percent for the most long-lived states.

IV. DISCUSSION

The results from the lifetime measurements are plotted in Figs. 4 and 5. For the lower states values are taken from Refs. 14, 7, 8, and 10. In an unperturbed Rydberg sequence the lifetimes are expected to be proportional to a power, close to three, of the effective principal quantum number. For the ${}^2S_{1/2}$ states, as can be seen in the log-log scale of Fig. 4, the values fall on a straight line, which demonstrates the unperturbed character of this sequence. From a

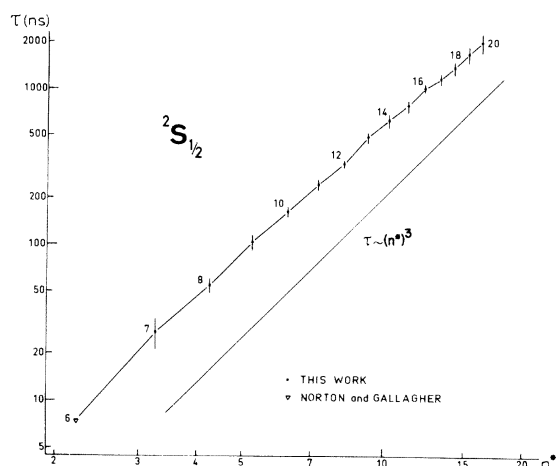


FIG. 4. Experimental lifetimes for the $5s^2ns^2S_{1/2}$ ($n=6-20$) states plotted vs the effective principal quantum number. A straight line showing the lifetime trend in the case of an $(n^*)^3$ dependence is also drawn. Our error bars are represented by the vertical lines.

fit we obtain $\tau = 0.955 n_{\text{eff}}^{2.77}$. The 2D sequences, however, show several strange features. For both the $^2D_{3/2}$ and $^2D_{5/2}$ states there is a drastic increase in lifetime values around $n=7-10$. There is also a large difference in lifetimes between the two fine-structure levels. For low n values the $^2D_{5/2}$ states have a shorter lifetime than the $^2D_{3/2}$ states, while for high n values the situation is reversed.

The effect of perturbations on lifetime values have been studied in several Rydberg sequences in the alkaline-earth elements (Ref. 1 and references therein). In these sequences short-lived states with two excited electrons mix into the energetically close-lying Rydberg levels and cause a decrease in the lifetimes. This effect can be used to determine the degree of configuration mixing. In the 2D sequences of indium, however, the situation is more complicated since no doubly-excited states exist in the energy range of the $(7-10)^2D$ states. In the measurements, the fluorescent light obtained from the atomic beam was one or two orders of magnitude weaker for these states than for the next higher levels. Low-transition probabilities have also been found in absorption measurements⁵ and a similar behavior persists in Ga and Tl. The low-absorption cross sections for these states in In have been sug-

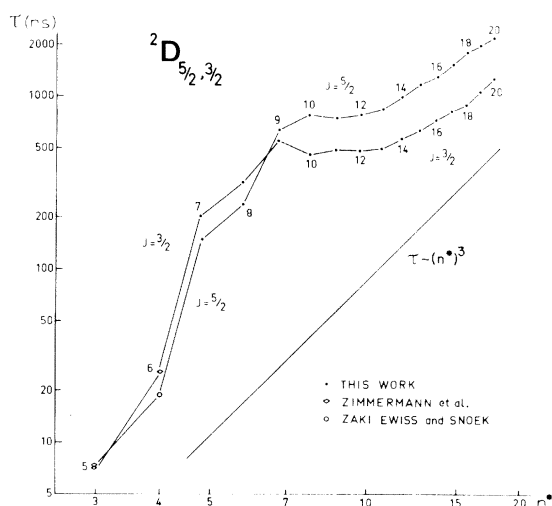


FIG. 5. Experimental lifetimes for the $5s^2nd^2D_{3/2,5/2}$ ($n=5-20$) states plotted vs the effective principal quantum number. Experimental results from earlier measurements are taken from Refs. 7, 8, and 10.

gested to be owing to configuration interaction with the autoionizing $5s5p^2^2D$ states.¹⁵ As can be seen in Fig. 5, the lifetimes increase monotonically for higher states but the lifetime values are different in the $^2D_{5/2}$ and $^2D_{3/2}$ sequences. This indicates that a perturbation strongly affects the levels around $n=7-10$ and in a more smooth way the rest of one or both sequences. Clearly, theoretical calculations including configuration-interaction effects are necessary to explain these results.

Lifetimes for the three $5s5p^2^4P$ states were also measured. In pure LS coupling these states would be metastable and the lifetime values are probably a measure of the degree of intermediate coupling. From a fit of energy levels no deviation from LS coupling is obtained, but for similar elements, e.g., Tl I, and Sn II, and $J = \frac{3}{2}$ have the highest purity and $J = \frac{5}{2}$ the lowest,¹⁶ as indicated by our lifetime results.

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