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Measurements of lifetimes of sodium Rydberg states in a cooled environment

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We have measured lifetimes of sodium s and d levels in the range $17 < n < 28$. Lifetime shortening due to blackbody radiation has been effectively eliminated by carrying out the measurements in a thermally isolated container cooled to 30 K. The individual lifetimes have been determined to 5% and agree well with calculations based on the Coulombic approximation. The validity of the calculation has been further tested using non-Coulombic methods. An $(n^*)^3$ scaling law has been obtained from the lifetimes and is accurate to 2.4%.

I. INTRODUCTION

Accurate measurements of the lifetimes of highly excited states can provide sensitive tests for calculations of radiative matrix elements, oscillator strengths, and polarizabilities. Furthermore, the development of experimental techniques needed for these measurements is helpful for investigating related phenomena such as population transfer due to blackbody radiation or photoionization, and to applications such as infrared detection.[1,2] Alkali-metal atoms have frequently served as theoretical and experimental test systems for these problems.[3,4,5] Although the alkali-metal atoms should be among the easiest to treat theoretically, predictions of radiative lifetimes for Rydberg states are by no means consistent: variations of 5—15% can be found in the literature. The experimental situation is even more confused. For levels below $n = 15$ lifetimes are known generally 5—10%, but for higher-lying states the accuracy rapidly diminishes. For levels with lifetimes greater than 6 μsec, the accuracy has not been better than 25%.

We present here measurements of the lifetimes of sodium s and d states in the range $17 \leq n \leq 28$, where the lifetimes vary between 4 and 24 μsec. We believe that the results are reliable to 5%. In addition, we describe calculational methods which give values that are in good agreement with the experimental results.

Blackbody radiative transfer can appreciably alter observed decay rates, as has been noted by several groups.[3,6—11] Corrections can be made for blackbody effects, but they generally introduce sizable uncertainties. An important innovation in these measurements is the use of a helium-cooled apparatus which minimizes the need for corrections due to radiative transfer. Blackbody effects can be observed by raising the temperature; at room temperature the decrease in the lifetime is as much as 40%. Measurements of blackbody radiative transfer rates will be published elsewhere.

II. APPARATUS AND PROCEDURE

Rydberg atoms were prepared in an atomic beam using stepwise pulsed laser excitation, and their number was monitored by pulsed field ionization. The time evolution of the excited population was observed by varying the delay time for the ionizing pulse. Although simple in concept, the method offers many opportunities for systematic errors. The main burden of these measurements lay in identifying and eliminating these errors.

Techniques for exciting and detecting Rydberg atoms have been described elsewhere,[12] and we shall emphasize here only those features which are important to the lifetime measurements. A thermal beam of sodium atoms was employed. To

minimize geometrical effects introduced by the motion of the atoms, the visible lasers were aligned counterpropagating with the atomic beam in the manner described by Fabre.[11] Atoms which pass out of the detection region during the observation time are replaced by atoms coming from further upstream. This technique allows observation of atoms with delay times of up to 25 μsec, corresponding to drift distances as long as 2 cm.

The sodium atoms were excited in two steps. The first step saturated the $3s \rightarrow 3p$ transition, and the second step, which was well below saturation, drove the $3p$ to Rydberg-state transition. Great care was taken to assure uniform excitation over the entire length of the drift region of the Rydberg atoms. Both laser beams were passed through pinhole apertures for spatial filtering, and focused at infinity. The atomic beam was collimated to a diameter of 1 mm with a divergence angle of 0.003 rad. The laser beams were collimated to diameters at least 50% larger than the atomic beam, assuring a uniform flux of atoms to the detection region.

The active region was centered between two parallel field plates, 7.5 cm in diameter and 1 cm apart. The field plates and surrounding thermal shields were attached to a helium dewar and this apparatus was in turn shielded by a liquid-nitrogen-cooled jacket. The temperature of the thermal shield was monitored by a carbon resistance thermometer. Independent measurements assured us that the field plates and the heat shield were in good thermal equilibrium. A heat leak prevented us from achieving temperatures below 30 K, but radiative transfer at 30 K is so small that the heat leak was not a serious problem. Further details on the thermal shielding will be published elsewhere.

III. DATA ANALYSIS

A number of small corrections are required to obtain the radiative lifetimes from the data. The field ionization signal includes electrons not only from the initial ns or nd state, but also from a few adjacent levels. In observing s-state lifetimes, $(n+1)p$ and np states were also collected. For d-state lifetimes, the nd, $(n+1)p$, np, and nf levels were all collected. The contribution of spontaneous decay to these adjacent channels is small, typically 1%, so that corrections to the observed lifetime could be made with negligible error. An added advantage of collecting electrons from the adjacent levels is that it removes effects of blackbody-induced transfer to first order. At 30 K, approximately 90% of this transfer occurs to these levels. Since the total blackbody transfer rate never exceeded 3% of the spontaneous decay rate, no further correction for the effect was required.

The data were also corrected for the effect of flow of fast-moving atoms from outside the excitation region. Corrections were only necessary for the longest-lived states and only for the "tail end" of their decay curves. Omission of the correction changed the lifetime of the longest-lived state by less than 1%, and the total correction introduced negligible error. There was no evidence of any background signal associated with the atomic beam.

Numerous experimental checks were made to as-

TABLE I. Experimental and theoretical lifetimes of sodium s and d states.

	s states		d states	
Level	$\tau_{expt}(\mu sec)$	$\tau_{theor}(\mu sec)$	$\tau_{expt}(\mu sec)$	$\tau_{theor}(\mu sec)$
17			4.46(22)	4.48
18			5.75(28)	5.31
19	7.42(17)	7.58	6.90(34)	6.25
20	8.9(4)	8.94	7.7(4)	7.29
21	11.3(6)	10.5	8.6(4)	8.43
22	12.2(7)	12.1	10.2(5)	9.69
23	14.5(8)	14.0	11.4(6)	11.1
24	16.6(9)	16.0	13.9(7)	12.6
25	18.6(10)	18.3	15.0(7)	14.2
26	21.2(12)	20.7	16.9(8)	16.0
27	23.8(12)	23.3	17.7(9)	17.9
28	24.9(12)	26.1		

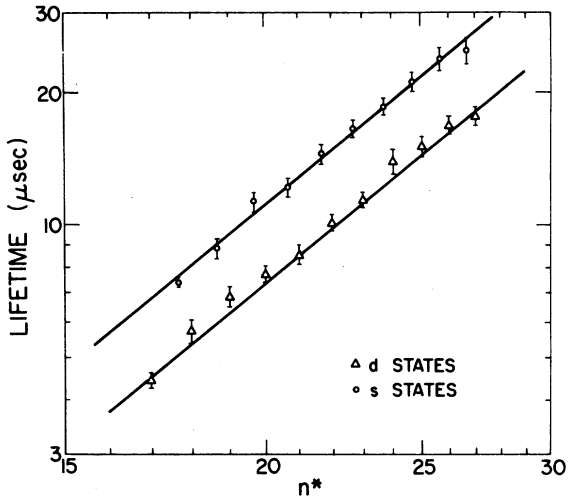

FIG. 1. Lifetimes of s and d states plotted versus principal quantum number.

sure that the data were not sensitive to the laser alignment, that they had the expected weak dependence on collimation, and that collisional or superradiant effects were absent. A small amount of transfer of d-state atoms to high angular momentum states was noted. We attributed this effect, which was comparable to the 30-K blackbody radiative transfer rate, to collisions with the background gas or ions accompanying the atomic beam. Since the states of high angular momentum were collected in the field ionization signal, errors due to this transfer were removed to first order.

Lifetimes were obtained from the data by analyzing each decay curve in two sections. The first, lasting one time constant, was fit to a simple exponential using a least-squares routine. These data, which carried the highest statistical weight and were least sensitive to systematic errors due to blackbody transfer or beam uniformity, were the principal source of the final results. Data for the second section were fit to an exponential plus terms involving transfer and decay in the adjacent levels, and were checked for consistency with the values from the first section. In all cases the two values agreed within statistical error, and there was no evidence of systematic deviations between the expected short- and long-term behavior.

IV. THEORY

The Coulomb approximation is frequently employed for calculating radiative matrix elements for Rydberg states. We have used it to calculate radiative lifetimes with a matrix-element routine created by Zimmerman, described elsewhere.[12] The quantum defects were taken from a recent compilation by Martin.[13] We tested the calculations by comparing our results with similar calculations for some lower-lying states of sodium by Lindgard and Neilsen,[14] and also with some results of Gounand,[15] based on the Bates-Damgaard method. The calculations were generally in good agreement, the differences never exceeding 3%.

In spite of the consistency of these various calculations, questions can be raised about the accuracy of any lifetime calculation based on the Coulomb approximation. The Coulomb approximation is known to be extremely accurate for calculating matrix elements between Rydberg states,[12] but it is not obvious that it should give accurate values for matrix elements between Rydberg states and low-lying states. Since the lifetimes of Rydberg states are dominated by optical-decay channels to low-lying states, any treatment based on the Coulomb approximation is suspect.

To test the effect of a non-Coulombic core potential, we recalculated the lifetimes, extending the radial integration inward to include the core region. We employed a Hartree-Slater program[16] to generate the core potential. The effect on the lifetimes of including a realistic core potential was

TABLE II. Error budget for several representative levels. Transfer errors are due to residual blackbody radiation induced transfer which modifies the decay shape of the data. Focus errors include beam-shape errors, both of which modify the uniformity of excitation along the pencil of atoms.

Level	20s (%)	27s (%)	17d (%)	26d (%)
Transfer	1.1	1.6	0.7	1.4
Focus	0.9	2.4	0.4	1.7
Statistics	5.0	5.2	4.0	4.2
Total error	5.2	5.9	4.1	4.6

TABLE III. Lifetimes of s and d states fit to the scaling law $\tau=\alpha(n^*)^3$.

	α_{expt}(nsec)	α_{theor}(nsec)
s	13.90(30)	13.70
d	9.59(25)	9.12

slight, with changes in the the lifetimes never greater than 1%. Thus, at the level of accuracy of 1%, it appears that the Coulombic approximation is adequate for our needs.

It is helpful to have scaling laws for the lifetimes of Rydberg states, though only in exceptional cases can they be rigorously justified. If the lifetime is dominated by decay to low-lying states, then it can be expected to scale as $(n^*)^3$, where n^* is the effective quantum number. We find that the lifetime of the sodium s and d Rydberg states obey the $(n^*)^3$ law closely, as the results shown in Fig. 1 demonstrate. In contrast, the p-state lifetimes in the range $15<n<30$ are given by $\tau=\alpha(n^*)^{3.11}$, with $\alpha=8.0$ nsec. We attribute this deviation from the $(n^*)^3$ law to the weak coupling of the p levels to the ground state due to the concentration of oscillator strength in the principal transition.

V. RESULTS

Final results are given in Table I. One data point 19s, was subsequently studied in great detail at 6 K, which accounts for its relatively high statistical accuracy.

An error budget for several representative levels showing the major sources of error is shown in Table II. Because the relative importance of different sources of error varies with n, errors are shown separately for short- and long-lived states. "Focus" designates the error arising from laser-beam divergence and inhomogeneous beam shape. "Transfer" designates error caused by modification of the decay curve by transfer to the adjacent states. The percentages given for these sources represent estimated limits of error. Although the uncertainties due to correction for systematic errors varied appreciably with the lifetime, in no case were they a major contributor to the final error.

TABLE IV. Error budget for fit of lifetimes to the $(n^*)^3$ scaling law. Error appears as uncertainty in the value of the scaling coefficient α.

	s levels (%)	d levels (%)
Transfer	1.4	1.2
Focus	1.4	1.1
Statistics	1.3	1.5
Total error	2.4	2.2

The experimental and theoretical results are shown in Fig. 1, and appear to be in good agreement. This is confirmed by fitting the data to the expected $(n^*)^3$ scaling law. The scaling law results are shown in Table III along with an error budget for the scaling coefficient shown in Table IV. Since all the data are fit to the scaling law the statistical error is reduced, and the dominant source of error is systematic.

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[1]T. W. Ducas, W. P. Spencer, A. G. Vaidyanathan, W. H. Hamilton, and D. Kleppner, Appl. Phys. Lett. 35, 382 (1979).

[2]H. Figger, G. Leuchs, R. Straubinger, and H. Walther, Opt. Commun. 33, 37 (1980).

[3]F. Gounand, M. Hugon, and P. R. Fournier, J. Phys. (Paris) 41, 119 (1980).

[4]T. F. Gallagher, S. A. Edelstein, and R. M. Hill, Phys. Rev. A 11, 1504 (1975).

[5]S. Nakayama, F. M. Kelly, and G. W. Series, J. Phys. B 14, 835 (1981).

[6]T. F. Gallagher and W. E. Cooke, Phys. Rev. Lett. 42, 835 (1979).

[7]E. J. Beiting, G. F. Hildebrandt, F. G. Kellert, G. W. Foltz, K. A. Smith, F. B. Dunning, and R. F. Stebbings, J. Chem. Phys. 70, 3551 (1979).

[8]G. F. Hildebrandt, E. J. Beiting, C. Higgs, G. J. Hatton, K. A. Smith, F. B. Dunning, and R. F. Steb-

bings, Phys. Rev. A 23, 2978 (1981).
[9]W. E. Cooke, and T. F. Gallagher, Phys. Rev. A 21, 588 (1980).
[10]J. W. Farley and W. H. Wing, Phys. Rev. A 23, 2397 (1981).
[11]C. Fabre, Ph.D. thesis, Paris, 1980 (unpublished).
[12]M. L. Zimmerman, M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. A 20, 2251 (1979).
[13]W. C. Martin, J. Opt. Soc. Am. 70, 784 (1980).
[14]A. Lindgard and S. E. Nielsen, At. Data Nucl. Data Tables 19, 533 (1977).
[15]F. Gounand, J. Phys. (Paris) 40, 457 (1979).
[16]F. Herman and S. Skillman, *Atomic Structure Calculations* (Prentice-Hall, Englewood Cliffs, N. J., 1963).