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Accurate lifetime measurements of the lowest $^2P_{1/2}$ states in neutral lithium and sodium

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By observing the decay in flight of laser-excited atoms in a fast atomic beam with energy between 60 and 150 keV, lifetimes of the Li $2^2P_{1/2}$ and Na $3^2P_{1/2}$ are measured to be (27.29 ± 0.04) and (16.40 ± 0.03) nsec, respectively (uncertainties are one standard deviation).

I. INTRODUCTION

Knowledge of spectra and energy levels of atomic systems has reached high standards. In contrast, the available information about lifetimes and transition probabilities is more limited and of less accuracy, but is of as basic scientific interest as the former.[1] In this paper we report on an experimental determination of two lifetimes with a relative uncertainty of less than 2×10^{-3}.

The principal significance of these high-accuracy measurements is the comparison to theoretical work. In calculating wave functions, electronic correlation can only be treated approximately. A variational approach determines wave functions most accurately in regions of space weighted most heavily by the Hamiltonian. In other regions wave functions depend sensitively on the approximations used, and even on computational details. Transition matrix elements weight different regions than the Hamiltonian. Therefore reliable lifetime data can furnish sensitive tests of wave functions.

Beyond the two-electron system,[2] accurate calculations of oscillator strengths are typically claimed to be reliable at the 5% level.[3] The experimental uncertainties as reported here can lead to meaningful conclusions only in "simple" systems. As the simplest, but nontrivial systems, Li and Na have to be considered.

There is a large amount of theoretical work on these systems, but the measurements, although numerous in Na, are not sufficiently accurate to distinguish between different theoretical approaches. These facts and experimental advantages have led us to measure lifetimes in Li and Na to an accuracy of a few parts in a thousand.

The first resonance line occurs in Li at 670.8 nm, and in Na at 589.6 nm. The fast beam laser technique (FBL) is applicable[4]: A fast neutral beam of typical speed 1 mm/nsec is produced by neutralizing the charged beam of an accelerator in a gas cell. Excited states are selectively populated by laser light irradiating the atoms at a well-defined point in space. The subsequent free decay in flight is detected as a function of distance. The time base and the time resolution are given via the speed of the particles and the spatial resolution of the detector, respectively (see Fig. 1 below and Refs. 5 and 6).

This technique shares the unperturbed decay with the standard beam-foil method.[7-9]. The selective excitation solves the cascade problem. The intense signals obtainable in FBL allow lifetime measurements better than 1% for the first time in atomic physics.[6]

II. EXPERIMENTAL SET UP

When performing a high-precision lifetime measurement, points of major concern are the linearity of the time base, instabilities of the set up, coherence effects, the intensities of the fluorescence signal and the background, and the calibration of the time base.

The time base is given by the speed of the radiating particles, and is affected by the velocity distribution in the beam.

The energy width of the charged beam emerging from the accelerator was measured to be less than 7×10^{-4} (see Sec. III below). Neutralizing collisions with thermal particles in a gaseous target[5] increase both the energy spread and the angular divergence. At a surface density of 10^{13} cm^{-2} of n hexane, no deterioration of the beam could be observed. This has been checked with a movable particle detector.[10]

A high vacuum gauge at the beam position in the

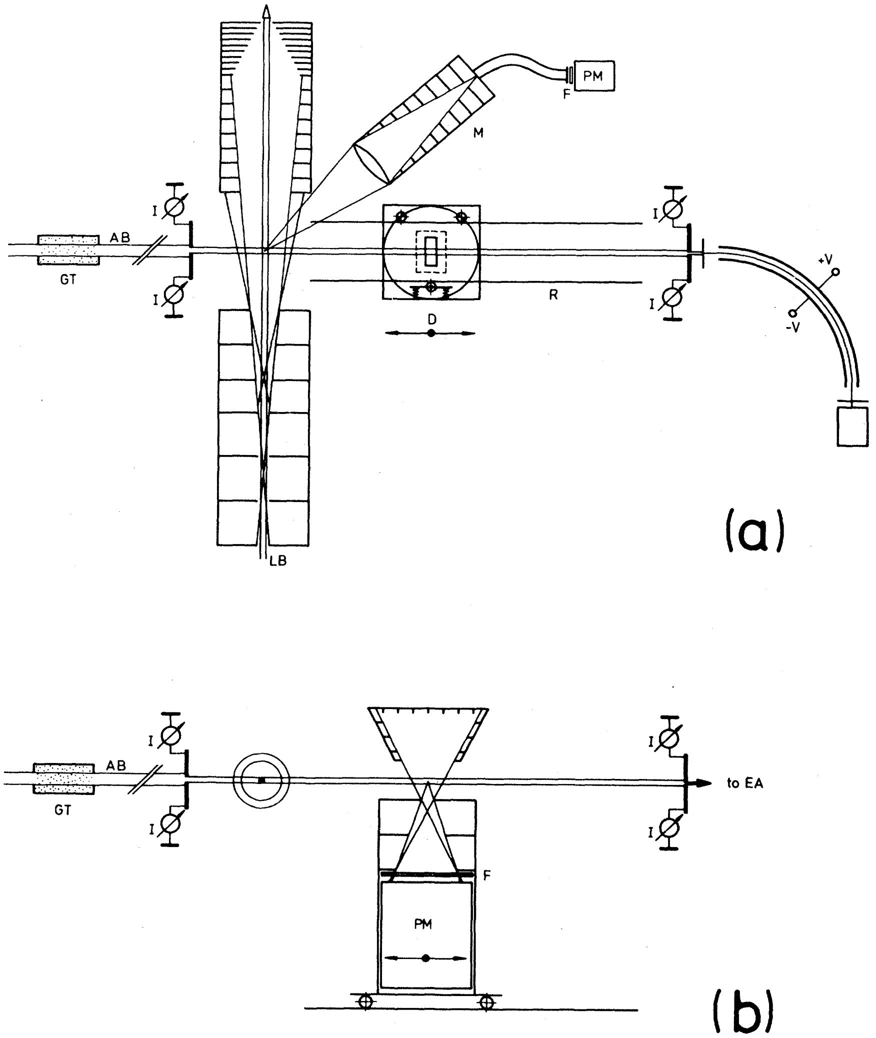


FIG. 1. Experimental set up, schematic. (a) Top view, (b) side view. AB: atomic beam; GT: cryogenically cooled gaseous target; I: beam defining apertures with current meters; LB: laser beam with stray light suppressing apertures; D: movable detector with spring-loaded wheels; M: monitor detector with lens, stray light suppression system, and flexible light guide; EA: adjustable electrostatic energy analyzer with entrance and exit slits, high-voltage supplies, and particle detector; F: optical filter; PM photomultiplier.

experimental chamber detected no thermal gas jet from the gas target under conditions of actual measurements.

The detector was mounted on a carriage riding on a rail inside the vacuum of the experimental chamber. Kinematical mounting ensured that the line of motion was given by the rail, which was a precision-machined optical bench. A stepping motor rotated a high-precision stainless steel screw, which in turn moved the carriage via a spring-loaded nut. The detector position was reproducible within 0.05 mm. The differential and integral linearity was better than 0.003 mm over any 30-mm distance according to the manufacturer of the screw. The actual pitch, however, was affected by thermal effects.

Two rectangular apertures of 1.5-mm height and 4-mm width separated by 500 mm on the rail defined the beam axis within ±0.5 mrad. They were made from four insulated razor blades each annealed to reduce magnetic fields. The current on these aperture was used to steer the beam.

The residual lateral motion of the detector with respect to the actual beam was less than ±0.2 mm over 200 mm, the longest travel used.

The detector was mounted below the beam. It contained a system of rectangular aperture limiting the angular range of accepted light. Below there was a wavelength filter and a photomultiplier (see Fig. 1).

The detection efficiency function was given by the solid angle with which the detector "saw" a given point in space assuming constant efficiency across the photocathode. This function was a triangle along the beam axis, and a trapezoid along a horizontal line perpendicular to the beam. The triangle had 8-mm full width at half maximum, and a geometrical analysis showed that the residual vertical beam displacement could affect the lifetime by up to $\pm 7\times 10^{-4}$.[11] The trapezoid had a roof length of 8 mm, wider by a factor of 2 than the beam. Any horizontal displacement of the beam had been shown experimentally to have a negligible effect on the collection efficiency. The solid angle was 8×10^{-3} sr.

The housing of the multiplier tube was thermally connected to a liquid air container via a flexible copper band. Dark signal was 20 to 40 cps at the discriminator level used.

Above the beam an open box painted black inside intercepted the solid angle of the detector. The photocathode was illuminated over a large area reducing the effect of efficiency variations.

Stray magnetic fields were compensated within $\pm 2\times 10^{-6}$ T at the beam. At the position of the phototube the shielding was less efficient, requiring a correction of the lifetime in Na of $(-2.5\pm 1)\times 10^{-4}$. For the Li experiment the field compensation was improved and a μ-metal shield was installed around the tube. No response variation was observed within counting statistics of $\pm 1\times 10^{-4}$.

For normalization a quantity proportional to the actual number of excited atoms must be known. Thus a second monitor detector observed light from a fixed part of the beam containing the region of excitation. Its optical axis was in the horizontal plane at approximately 45 degrees with respect to both the atomic and laser beam axis. The latter was horizontal and perpendicular to the former one. The longer distance necessary to stay clear from the detector was provided by a lens of $f=30$ mm and 30 mm diameter focusing light onto a flexible light guide. At its other end was the vaccum window, a wavelength filter, and a photomultiplier (see Fig. 1.) Extensive testing showed the monitor signal to be independent of detector position.

A jet stream dye laser delivered up to 0.5 W at a linewidth of 3 GHz near 589.6 nm, and up to 0.2 W at a linewidth of 0.1 GHz near 670.8 nm.[12] The Gaussian beam was made to cross the atomic beam at 90° in the horizontal plane with linear horizontal polarization. The laser was focused to less than 1 mm diameter.

The observed excitation probability of 5% compared well with the theoretical estimate[11] of 10% given the difficulties in estimating the neutral particle current. The observed resonance width was 5 GHz in Na and 3 GHz in Li. The smaller latter value was due to single-mode operation of the laser.

The detected signals contained not only true fluorescence light, but also contributions due to parasitic laser light ("stray light"), collisional excitation ("beam noise"), and noise of the detector ("multiplier noise"). This background constitutes a major problem in precision lifetime measurements. The following measures were taken to reduce it experimentally.

Both multipliers were cooled to below −10° C.

Laser stray light was reduced by a system of circular apertures around the laser beam extending 450 mm on either side of the atomic beam.[11] Stray light levels below 1000 cps per Watt laser power were achieved.

The experimental chamber was evacuated by turbomolecular pumps at a speed of 200 l/sec, and by liquid-nitrogen-cooled surfaces. Routine pressure readings were 2×10^{-6} mbar. Collisional excitation gave a signal of 1000 cps per μA beam current.

Collisions in the gas target populated long-lived states cascading into the level under investigation. At a distance of 1.5 m between excitation and detection, this background signal exceeded 10 000 cps, and had a slope corresponding to a mean life of some 100 nsec. The true fluorescence signal of 3×10^{5} cps at maximum intensity before dead-time correction compared favorably with these backgrounds.

In order to determine the remaining backgrounds as realistically as possible, a digital lock-in technique was adopted chopping the laser and the atomic beam at each position of the detector.[5,13] No data were taken before everything had settled from the changes made. Different time intervals were used for the four combinations, laser beam on/off, atomic beam on/off. For a particular run the detector moved up and down the rail 20 times, adding to all previous sweeps of that run. Data were stored separately for later treatment.

The accuracy by which a decay constant can be determined during a given measuring time depends on the length along the decay curve over which data are taken. Numerical simulations showed an optimum length of three to six mean decay lengths at

TABLE I. List of systematic errors (in 10^{-4})

Linearity test of detector	-2.5 ± 1		±1
Dead-time correction	±3		±3
Aligment of atomic beam and detector movement	±7		±7
Stopping in gas target	$+8\pm8$		±8
Collision with rest gas	$+4.5\pm3$		±3
Quantum beats	±3		±3
Energy measurements	±3		±3
Calibration factor of energy analyzer		±7	
Thermal expansion of driving screw		±3	
Statistics	±7		±7
Addition in quadrature	$+10\pm16$		±16

actual background situation.

Data treatment consisted of a dead-time correction of a few percent allowing for a 30% fluctuation in the mean counting rate as estimated from the experimental instabilities. After background subtraction and normalization with respect to the monitor fluorescence signal, a single exponential is fitted by the standard least-squares method.

III. VELOCITY DETERMINATION

The most direct and most general method would be to measure the pitch of the screw and the Doppler effect of the laser-excited atoms. However, at a required resolution of 1 GHz signal levels are low. Therefore an electrostatic energy analyzer was used. As in earlier work,[5,13] the analyzer was calibrated against the He I 3^3P $J=1$ to 2 fine-structure splitting,[14] which gives rise to easily observable quantum beats after beam-foil excitation. Improvements of the calibration procedure led to an uncertainty of a suitably defined calibration factor of $\pm7\times10^{-4}$.[15] This uncertainty was dominated by the assumption that the center of the energy spectrum after foil interaction can only be determined within $\frac{1}{20}$ of the linewidth. Thermal effects could change the actual pitch of the screw. Its fractional variation from the lifetime measurements to the calibration was less than 3×10^{-4}.

IV. SYSTEMATIC ERRORS

Operating the laser 0.4 nm off resonance, the signal in 39 out of 64 detector positions was compatible with zero within one standard deviation after background subtraction, as is expected from a normal distribution. Deliberately steering the atomic beam off axis, the error in lifetime was conservatively estimated to be less than $\pm7\times10^{-4}$, in agreement with predictions (Table I).

At an ambient pressure in the experimental chamber above 10^{-5} mbar, the Na decay curve significantly deviated from an exponential. At the usual pressure a fractional correction of $(+4.5\pm3)\times10^{-4}$ applied to the lifetime. Owing to possible gradients we only give the order of magnitude for the cross section of depopulation to be 10^{-14} cm^2. Extrapolation to zero pressure in the gas target led to a relative correction of $(+8\pm8)\times10^{-4}$ at normal operation conditions. For Li no effect was observable within experimental

TABLE II. Comparison of lifetime measurements in Li 2^2P.

Ref.	Method	τ (ns)	f value
33	Level crossing	27.2 ± 0.4	0.744 ± 0.011
20	Beam gas	31.9 ± 1.6	0.63 ± 0.03
34	Beam foil	25 ± 1	0.81 ± 0.03
35	Beam foil	26.2 ± 1.0	0.77 ± 0.03
	FBL this work	27.29 ± 0.04	0.7416 ± 0.0012

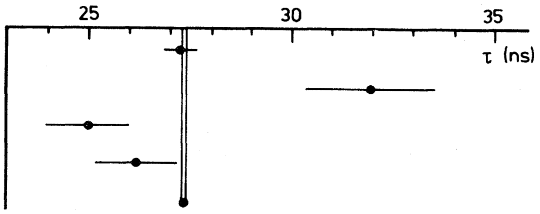
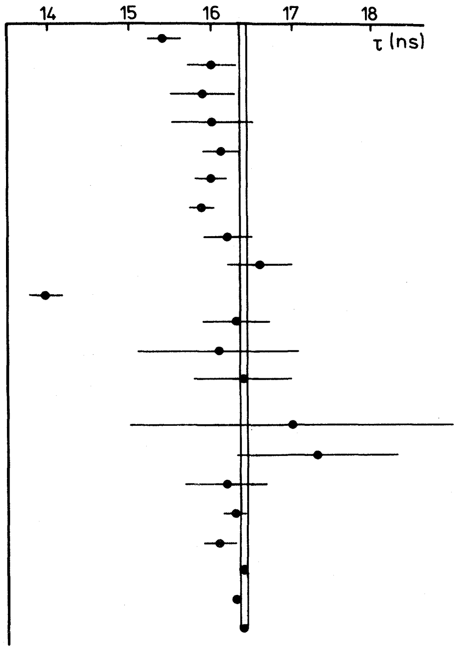


TABLE III. Comparison of lifetime measurements in Na 3^2P.

Ref.	Method	τ (ns)	f value[a]
21	Hook method	15.4 ± 0.2	1.014 ± 0.013
36	Level crossing $J=\frac{3}{2}$	16.0 ± 0.3	0.976 ± 0.018
37	Level crossing $J=\frac{3}{2}$	15.9 ± 0.4	0.982 ± 0.025
38	Level crossing $J=\frac{3}{2}$	16.0 ± 0.5	0.976 ± 0.030
39	Level crossing $J=\frac{3}{2}$	16.12 ± 0.22	0.969 ± 0.013
40	Hanle effect $J=\frac{3}{2}$	16.0 ± 0.2	0.976 ± 0.012
22	Phase shift	15.9 ± 0.16	0.982 ± 0.010
41	Phase shift	16.2 ± 0.3	0.964 ± 0.018
42	Phase shift	16.6 ± 0.4	0.941 ± 0.023
43	Pulsed electron beam	14.0 ± 0.2	1.115 ± 0.016
44	Pulsed optical excit.	16.3 ± 0.4	0.958 ± 0.024
45	Pulsed optical excit.	16.1 ± 1.0	0.970 ± 0.060
46	Pulsed optical excit. $J=\frac{1}{2}$	16.4 ± 0.6	0.952 ± 0.035
47	Beam-foil	17 ± 2	0.92 ± 0.11
48	Beam-foil	17.3 ± 1.0	0.90 ± 0.05
49	Beam-foil	16.2 ± 0.5	0.96 ± 0.03
50	FBL $J=\frac{1}{2}$	16.3 ± 0.16	0.959 ± 0.009
50	FBL $J=\frac{3}{2}$	16.1 ± 0.2	0.969 ± 0.012
23	FBL $J=\frac{1}{2}$	16.396 ± 0.026	0.9536 ± 0.0015
23	FBL $J=\frac{3}{2}$	16.339 ± 0.039	0.9550 ± 0.0023
	FBL $J=\frac{1}{2}$ this work	16.40 ± 0.03	0.9536 ± 0.0016



[a] Conversion to f values excluded the statistical weight to make values comparable.

accuracies.

Quantum beats[16–18] could degrade the decay curve requiring an elaborate data analysis. However the fine structure was resolved while the hyperfine structure was not. Also circularly polarized light components were suppressed. Therefore zero-field quantum beats could not occur. Relating the Zeeman matrix element to the mean decay rate, an upper limit of the uncertainty in the lifetime due to magnetic-field-induced quantum beats of $\pm 3\times 10^{-4}$ was estimated. After velocity calibration the 18 measurements in Na scatter over $\pm 15\times 10^{-4}$, matching the uncertainty of an individual run. The mean was taken to have a statistical uncertainty of $\pm 7\times 10^{-4}$. In Li no isotope effect was found. The statistical situation of 23 measurements was similar.

V. RESULTS

Taking the systematic corrections and uncertainties into account, we obtained for the lifetimes

$$(\mathrm{Li}\,2^2P_{1/2})=(27.29\pm 0.04)\ \mathrm{nsec}\ ,$$

$$(\mathrm{Na}\,3^2P_{1/2})=(16.40\pm 0.03)\ \mathrm{nsec}\ .$$

Quoted errors are one standard deviation. Using the experimental wavelength of the $D1$ line, the absorption oscillator strength is

$$f(\mathrm{Li}\,2^2S_{1/2}-2^2P_{1/2})=\tfrac{1}{3}\times(0.7416\pm 0.0012)\ ,$$

$$f(\mathrm{Na}\,3^2S_{1/2}-3^2P_{1/2})=\tfrac{1}{3}\times(0.9536\pm 0.0016)\ .$$

Identifying the numbers in parentheses with the total multiplet oscillator strength involves the assumption of statistical distribution. While this is likely to be valid in Li, a recent measurement by Gawlik *et al.*[19] seems to indicate deviations in Na. Using their relative f values, we obtain

$$f(\mathrm{Na}\,3^2S_{1/2}-3^2P_{3/2})=\tfrac{2}{3}\times(0.9465\pm 0.0023)\ .$$

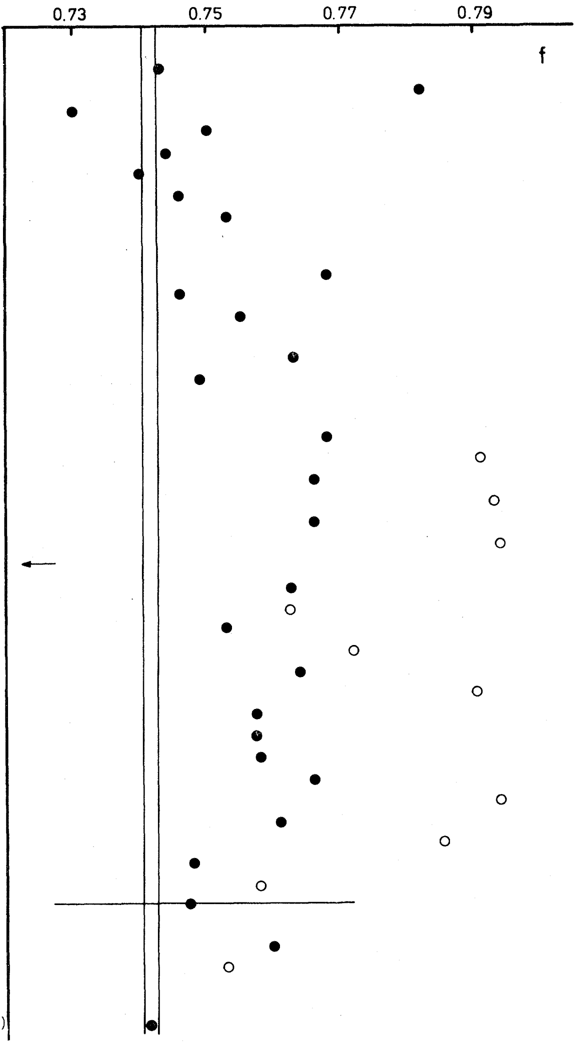
VI. DISCUSSION

A. Comparison to experiments

In Tables II and III we compare the experimental results available in the literature. Owing to greatly improved error limits, any disagreement with our data should be attributed to an underestimate of errors in those experiments. Within twice the quoted errors there is general agreement with only a few exceptions.

TABLE IV. Theoretical calculations of oscillator strength in Li 2^2S-2^2P.

Ref.	Method	f value[a]
	(a) Semiempirical calculations	
51	Coulomb approximation	0.743
52	Effective n^* and l^*	0.782
53	Scaled Thomas-Fermi ion potential	0.730
54	Scaled Thomas-Fermi pot. with pol.	0.750
55	Hartree-Fock core	0.744
56	Hartree-Fock core with polarization	0.74
57	Model potential with pol. and retard.	0.746
58	Adj. exchange potential	0.753
	(b) *Ab initio* Potential calculations	
59	Pseudopotential	0.768
60	Exchange pot. with polarization	0.746
61	Effec. pot. with energy-dependent polarization	0.755
27	Transition states in exchange potential	0.7629
62	Dirac Eq. in ex. pot. with polarization	0.749
	(c) SCF calculations	
63	Analytic Hartree-Fock	0.768
		0.791
	Numerical Hartree-Fock (This work)	0.766
		0.793
64	Frozen-core Hartree-Fock	0.766
		0.794
65	HF with core pol.	0.71
66	Pol. by effective operator	0.7625
		0.7626
63	45-term CI	0.753
		0.772
67	Spin-optimized SCF	0.7637
		0.7906
68	HF with freq.-dependent polarization	0.7577
69	Time-dependent Hartree-Fock	0.7575
70	Random-phase approx. with exch.	0.758
28	Transition states in HF	0.7666
		0.7943
71	Multiconf. Dirac HF	0.766
		0.788
72	Hylleraas wave function	0.748
		0.758
73	Hylleraas wave function with upper and lower bound $0.7227 < f < 0.7720$	0.7476
	FOTOS (this work)	0.760
		0.753
	(d) Experiment	
	FBL (this work) see text	1/3(0.7416±0.0012)



[a] Two entries correspond to length (●) and velocity (○) form, respectively.

Li. The beam gas lifetime measurement by Buchet *et al.*[20] gives too long a lifetime, the reason being probably cascades and pressure gradients and/or the use of an ion beam not analyzed with respect to mass.

Na. To arrive at an absolute number for the f value from a measure of the anomalous dispersion (hook method),[21] the number of scatterers in the active volume must be known, and the problems in this determination could explain the discrepancy.

We consider as more serious the deviation from the phase-shift measurement.[22] Coherence effects due to hyperfine interaction not considered by the author lead to a correction of -0.1 nsec, thus surmounting the difference. Exceptionally good is the agreement with the fast beam laser experiment of Schulze-Hagenest.[23] Not only is a slightly smaller uncertainty reported, but also the $J=\frac{3}{2}$ lifetime is determined with comparable uncertainty in spite of the superimposed quantum beats.

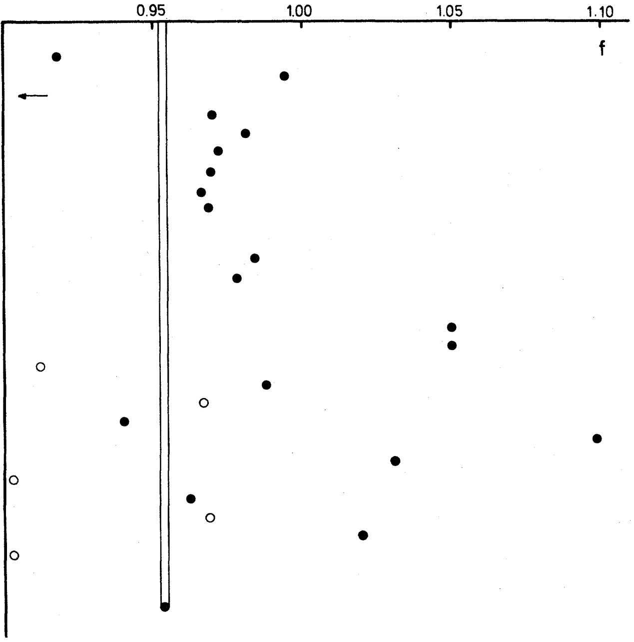
B. Comparison to theory

In Tables IV and V, our results are compared to calculations of oscillator strength. These are ordered with respect to increasing complexity within each subgroup. In view of the discussions of which form of oscillator strength is to be used (Refs. 24–26 and references therein), both length and velocity forms are given where appropriate and available. We follow the authors' choice between experimental and theoretical energy difference.

Semiempirical calculations give on the average

Table V. Theoretical calculations of oscillator strength in Na 3^2S-3^2P.

Ref.	Method	f value[a]
	(a) Semiempirical calculations	
51	Coulomb approximation	0.918
52	Effective n^* and l^*	0.994
74	Effective l^*	0.75
53	Scaled Thomas-Fermi ion potential	0.97
54	Scaled Thomas-Fermi pot. with pol.	0.981
75	Hartree-Fock core	0.972
76	HF core with polarization	0.9694
58	Adj. exchange pot.	0.9659
77	Adj. exchange pot.	0.969
	(b) *Ab initio* potential calculations	
59	Pseudopotential	0.984
27	Transition states in exchange potential	0.9783
	(c) SCF calculations	
78	Analytic Hartree-Fock	1.05
79	Numerical Hartree-Fock	1.05
		0.912
80	Frozen-core Hartree-Fock	0.988
		0.967
65	Frozen-core HF with polarization	0.94
81	HF with core pol.	1.0882
66	Pol. by effective operator	1.031
		0.9028
82	Multiconf. HF	0.962
		0.969
	FOTOS (this work)	1.02
		0.903
	(d) Experiment	
	FBL (this work) see text	1/3(0.9536±0.0016)



[a] Two entries correspond to length (●) and velocity (○) form, respectively.

smaller f values than *ab initio* methods and thus *better agreement with experiment.* Including correlation over the Hartree-Fock approximation yields smaller f values decreasing the deviation from experiment.

Surveying the literature, it seems unlikely to make significant further progress by "brute force"-type approaches such as systematically extending the length of CI expansions. An interesting idea is to treat both states of a transition simultaneously. This idea has not yet been followed very far, and the available results[27,28] are discouraging. We therefore decided to perform some calculations using an idea put forward some time ago by Nicolaides and Beck called the "first order theory of oscillator strength" (FOTOS, Ref. 29). It provides rules to select configurations in a CI expansion. This selection is done in view of the other state of the transition. The calculation of the wave functions is done for each state at a time. Since configurations are not selected on an energy basis we used experimental energy differences. A nonrelativistic CI code written by Beck and Nicolaides[30] was employed.

FOTOS gave a significant improvement over the Hartree-Fock approximation in the sense that the difference between the length and velocity forms decreased, and the agreement with experiment became better. Going beyond the first order vastly increased the computing time with little or no further improvements. This could be traced back to excluding energetically important configurations, while at the same time including some obeying Brillouin's theorem.[31] The latter ones are then poorly defined while making by construction important contributions to the f-value integrals.

VII. CONCLUSION

By using selective optical excitation of a fast atomic beam (fast beam laser technique), we measured lifetimes of the lowest excited states in neutral lithium and sodium with an uncertainty of less than two parts in a thousand (one standard deviation). The cases studied are experimentally favorable and theoretically simple. The experimental accuracy can be used to differentiate for the first time between elaborate calculations of oscillator strength. The theoretical literature on these transitions shows a trend to smaller f values when improving wave functions. Our results serve as limiting values for this trend. If this trend is a general feature also for transitions between other types of configurations, the often-quoted systematic discrepancy between theoretical and beam-foil data[8,32] may need a reexamination; the f values derived from beam-foil measurements are usually smaller than corresponding theoretical values.

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