Precision Lifetime Measurements on NaI $3p\ ^2P_{1/2}$ and $3p\ ^2P_{3/2}$ by Beam-Gas-Laser Spectroscopy

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The lifetimes of the fine-structure levels $3p\ ^2P_{1/2}$ and $3p\ ^2P_{3/2}$ in neutral sodium were determined at improved precision ($\pm 0.13\%$, 1σ) by means of the beam-gas-laser spectroscopy method. The resulting lifetimes of 16.299(21) and 16.254(22) ns, respectively, are in excellent agreement with the results of the most refined theoretical calculations. The long-standing discrepancy between *ab initio* line strength calculations and the measurements of Gaupp *et al.* [Phys. Rev. A **26**, 3351 (1982)] for the NaI $3s$-$3p$ transition appears to be resolved now. In addition, the hyperfine constants of the $3p\ ^2P_{3/2}$ level were determined.

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For more than ten years there has been a persistent discrepancy between *ab initio* theoretical line strengths for the resonance transitions in the light alkali atoms lithium and sodium and the experimental results published by Gaupp, Kuske, and Andrä [1] in 1982. These authors used the newly developed method of beam-gas-laser spectroscopy (BGLS) to determine the lifetimes of the first excited states in lithium and sodium and extracted line strengths for the transitions LiI $2s$-$2p$ and NaI $3s$-$3p$ with quoted uncertainties of $\pm 0.16\%$ only. A previous preliminary report on independent BGLS lifetime measurements on sodium by Schmoranzer, Schulze-Hagenest, and Kandela [2] was in agreement with Gaupp's result. However, the line strengths obtained from extensive *ab initio* calculations are consistently larger than the above experimental ones by 0.6% or more for both lithium and sodium. This discrepancy is particularly difficult to understand in the three-electron system lithium which is simple enough to allow for an almost exact theoretical treatment (for a review see Brage, Fischer, and Jönsson [3]). The *ab initio* calculation of line strengths in sodium is certainly a much more difficult task. Nevertheless, a few large-scale calculations have been reported recently (Guet, Blundell, and Johnson [4], Brage, Fischer, and Jönsson [3], and Jönsson *et al.* [5]) which can claim an accuracy of $\pm 0.3\%$ [4] or even better. The line strength results from these *ab initio* calculations are in a good mutual agreement but differ from the experimental result of Gaupp, Kuske, and Andrä [1] by $+0.6\%$ to $+0.9\%$. Only a rather simple semiempirical model-potential calculation (Theodosiou and Curtis [6]) reproduced Gaupp's result. Another more recent semiempirical calculation (Laughlin [7]), however, gave a result closer to the *ab initio* results mentioned above.

Therefore the lifetimes of the $3p$ fine-structure levels in sodium were measured again at improved precision by beam-gas-laser spectroscopy. The experimental technique has been described previously in more detail [8,9]; therefore only the essential features will be given here. The experimental setup was improved further in order to reach the desired accuracy level below $\pm 0.2\%$. Briefly, a fast monoisotopic ion beam is partially neutralized by charge exchange collisions in a gas cell and subsequently excited by a tuned cw dye laser. The emitted fluorescence photons are observed by means of a movable detector as a function of the distance from the laser intersection point. Hence, one obtains an exponential decay curve in space. The lifetime of the excited state is then determined from the spatial decay constant and the beam velocity.

In the present experiment a Na^+ beam was extracted from an ion-conductor source and accelerated to an energy of 170 keV. Argon was chosen as target gas for the charge exchange cell. At an operating pressure of 8×10^{-2} mbar about 30% of the ions were neutralized. The remaining ions were deflected out of the beam. The profiles of the atom beam were determined at several locations in the beam line by means of movable tungsten wires or movable slits.

After a free flight path of 2.3 m, which corresponds to a flight time of about 2 μs, the atom beam passed perpendicularly through the resonator of a linearly polarized cw dye laser which was tuned to one of the resonance transitions in sodium. The movable fluorescence detector consisted of 16 light-guide fiber bundles mounted axially symmetric around the atom beam. The cone-shaped detection head was translated parallel to the beam by a precision spindle drive. The light-guide bundles transmitted the collected photons to a monochromator and a cooled photomultiplier outside of the vacuum chamber. A similar fixed light-guide detection system served as a monitor to normalize the signal of the movable system.

Most of the laser stray light was separated from the fluorescence radiation by the monochromators due to the Doppler shift caused by the observation angles of 54.7° and 120° of the detector and the monitor, respectively. The remaining background signals were determined by means of laser and atom-beam choppers and subtracted automatically. Furthermore, the individual count rates were corrected for saturation effects of the detection system. The

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sum of all these corrections did not exceed 3% of the total signal at any position of the movable detector. The true fluorescence signal was about 100 000 cps at the first data point of the decay curves recorded.

A total of 21 decay curves, each consisting of 30 equidistant data points, was recorded for each of the two Na $3p$ fine-structure levels at a residual gas pressure of 8×10^{-7} mbar (see Fig. 1). A few additional runs were recorded at higher residual gas pressures in order to determine the correction for quenching. The data acquisition covered a flight path of 58 mm corresponding to about three lifetimes. The normalized intensity at the first point of the decay curves was constant within statistical uncertainty over all runs, thus precluding long-term drifts in the detection efficiency of the photon detectors.

The excitation with linearly polarized light ensures the absence of quantum beats in the decay curve of the $3p\ ^2P_{1/2}$ level. In this case a linear function was adapted to the logarithmized decay curves by means of a weighted least-squares fit. On the contrary the linearly polarized excitation of the $3p\ ^2P_{3/2}$ level generates an alignment which makes quantum beats in the decay curves of this level possible. The magic angle geometry of the detection system suppresses quantum beats completely as long as the transmission wavelength of the detector monochromator is tuned exactly to the Doppler-shifted wavelength which corresponds to the emission angle of 54.7°. With a small detuning of the monochromator, however, the effective observation angle can be shifted slightly. This degree of freedom was used in the present work in order to obtain some additional information about the hyperfine splitting constants of the $3p\ ^2P_{3/2}$ level. The observed hyperfine quantum-beat modulations of the decay curves had a peak-to-peak amplitude of 2.3% of the total signal. The logarithmized decay curves were evaluated by means of a weighted least-squares fit of the corresponding nonlinear theoretical expression (Fano and Macek [10]) which contains the hyperfine splitting constants A and B as additional fit parameters. Our results for the hyperfine constants are listed in Table I together with a few selected experimental and theoretical results from literature. The fit residuals (see Fig. 1) of both fine-structure levels were analyzed subsequently for deformations in the decay curves by a polynomial fit. The distribution of the quadratic and cubic coefficients was statistically consistent with zero, and a variation of the decay constants with the quadratic and cubic coefficients was not observed.

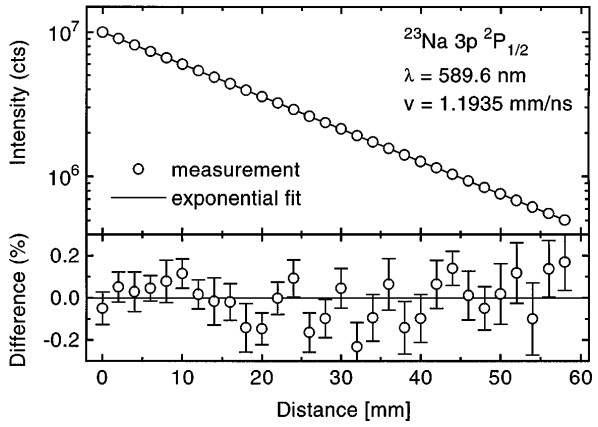

FIG. 1. Semilogarithmic plot of superposition of 21 measured decay curves of NaI $3p\ ^2P_{1/2}$ (upper part) and distribution of the intensity fit residuals (lower part).

The beam velocity was determined from the kinetic energy of the beam particles. The total acceleration voltage of the ion beam was measured to 169.94(12) kV by means of a calibrated voltage divider. The dependence of the ion beam energy on the operating conditions of the ion-conductor source used in the present work was investigated by means of an electrostatic energy analyzer. A small negative shift of the ion beam energy (with respect to the total acceleration potential) was observed, which amounted to $-220(15)$ eV under the operating conditions during the lifetime measurements. An upper limit of -50 eV for the energy loss caused by the neutralizing collision was determined by an energy analysis of the beam particles reionized in a second gas cell. The energy loss caused by the neutralizing collision was therefore estimated to $-25(10)$ eV.

TABLE I. Selected theoretical and experimental hyperfine constants for the NaI $3p\ ^2P_{3/2}$ level (uncertainties given in parentheses).

Ref.	Method	A (MHz)	B (MHz)
	Theoretical		
[11]	Coupled clusters	18.314[a]	...
[5]	MCHF+CI	18.80[a]	...
	Experimental		
[12]	Quantum beats (QB), BGLS	18.64(6)	2.77(6)
[13]	QB, pulsed laser	18.69(6)	2.83(10)
[14]	QB, pump/delayed probe	18.534(15)	2.724(30)
[15]	Laser spectroscopy	18.8(1)	2.7(2)
This work	QB, BGLS	18.79(12)	2.75(12)

[a]Corrected for relativistic effects (+0.06 MHz) using the ratio between Dirac-Fock (DF) and Hartree-Fock (HF) values.

Although the divergence angle of the atom beam is rather small in our BGLS experiment [approximately 0.7 mrad (FWHM)] the high precision desired for the lifetime results makes the beam divergence an important systematical error source. The beam divergence effect arises from the fact that the collection efficiency of the light-guide detector which collects the fluorescence photons after the laser excitation is not a constant in space but decreases with the radial distance from the beam axis [9]. As the detector is moved downstream along the beam in a lifetime measurement, the fraction of particles passing through the off-axis part of the detection region increases slightly due to the divergence of the beam. Hence one obtains an apparently faster decay of the fluorescence of the excited state, i.e., an apparently shorter lifetime. Our method to correct for the divergence effect was described previously [9]. The general relation between the apparent lifetime τ_a and the true lifetime τ_0 in the case of the observed linear beam divergence is

$$\frac{1}{\tau_a} = \frac{1}{\tau_0} + v_b C_{\mathrm{div}}, \tag{1}$$

where v_b is the beam velocity and C_{div} is a constant which is a complicated function of the beam profiles and of the detection geometry. In our BGLS setup the beam profiles and the spatial photon collection efficiency of the movable detector can be described to a good approximation by Gaussian-shaped model functions which leads to an expression for the constant C_{div} of

$$C_{\mathrm{div}} = \frac{w_x}{w_x^2 + w_d^2}\,\delta_x + \frac{w_y}{w_y^2 + w_d^2}\,\delta_y, \tag{2}$$

where w_d is the measured width of the detection region [9], w_x, w_y are, respectively, the horizontal and vertical widths (FWHM) of the atom beam at the start position of the decay curve, and δ_x, δ_y are the corresponding divergence angles. In principle, the beam parameters in Eq. (2) refer to the excited fraction of the atom beam. The exciting laser intersected the atom beam in horizontal direction; therefore the horizontal widths of the excited beam were identical to the widths of the full atom beam. The vertical widths of the excited beam, however, were found to be smaller than the widths of the full beam because of the small cross section of the laser beam. Inserting the measured parameters for the excited fraction of the atom beam into Eq. (2) the divergence constant becomes $0.18(4)\ \mathrm{m}^{-1}$ which leads to a beam divergence correction of the lifetimes of $+0.35(8)\%$.

The beam profile monitors which were mounted on the detector carriage for the determination of the beam divergence were also used to improve the alignment of the axis of the detector motion parallel to the atom beam. The uncertainty in the lifetimes due to detector disalignment was reduced down to $\pm 0.05\%$.

The total uncertainties of the lifetimes of the NaI $3p$ levels of about $\pm 0.13\%(1\sigma)$ result from a quadratic summation of the statistical and systematical uncertainties which are compiled in Table II. The uncertainties of the hyperfine constants are almost entirely statistical, with a small contribution of $\pm 0.04\%$ for the uncertainty in the beam velocity.

TABLE II. Amounts of corrections and uncertainties for the lifetimes of the NaI $3p\ ^2P_J$ fine-structure levels (in %).

	Corrections	Uncertainties (1σ)
Fit statistics		± 0.05
Dead time effects		± 0.03
Collisional quenching	$+0.016$	± 0.003
Beam velocity		± 0.04
Beam divergence	$+0.35$	± 0.08
Detector drive, adjustment		± 0.05
Detector drive, linearity		± 0.03
Light guide transmission		± 0.04
Zeeman quantum beats		± 0.03[a]

[a] $J = 3/2$ only.

Table III shows a compilation of experimental and theoretical line strengths for the Na $3s$-$3p$ transition, including a number of previously unpublished new results. The perfect accord among the three most recent very accurate experimental values, obtained by three completely different methods, is most remarkable.

Tiemann, Richling, and Knöckel [17] investigated the vibrational spectrum of the $A\ ^1\Sigma_u^+$ state of Na_2 by means of two-step Franck-Condon pumping of Na_2 molecules in a beam. The dominant interaction at large distances for this molecular state is of the resonant dipole-dipole

TABLE III. Recent theoretical and experimental lifetimes τ for NaI $3p\ ^2P_{1/2}$ and $^2P_{3/2}$ and total line strengths $S(3s$-$3p)$ (uncertainties given in parentheses).

Ref.	Method	J	τ_J (ns)	S (a.u.)
	Theoretical			
[6]	Semiempirical			37.03
[7]	Semiempirical			37.19
[4]	RMBPT all orders			37.38(11)
[11]	Coupled clusters			37.56[a]
[3]	MCHF-CCP			37.30[a]
[5]	MCHF+CI			37.26[a]
	Experimental			
[1]	BGLS	1/2	16.40(3)	37.04(7)[b]
[16]	Pulsed laser	1/2	16.35(6)	37.15(14)[b]
[17]	C_3 analysis	1/2	16.31(6)	37.24(12)[b]
[18]	Linewidth	3/2	16.237(35)	37.30(8)[b]
This	BGLS	1/2	16.299(21)	37.26(5)[c]
work		3/2	16.254(22)	

[a] Corrected for relativistic effects (-0.09 a.u.) using the ratio between DF and HF values. The original value of Ref. [3] without relativistic correction is 37.39 a.u.

[b] A line strength ratio between the two fine-structure components of 0.5 was assumed in the calculation.

[c] The ratio of the line strengths of the two fine-structure components was determined to 0.50014(44). This is in excellent agreement with the nonrelativistic prediction of 0.5. In the uncertainty estimate for the ratio all those systematical effects were omitted which affect both lifetimes in the same way.

type; therefore the asymptotic behavior of the internuclear potential can be described as C_3/r^3. The long-range interaction coefficient C_3 which is $1/2$ of the line strength of the $3s$-$3p$ transition (King and Van Vleck [19]) was extracted with an accuracy of $\pm 0.33\%$.

Oates, Vogel, and Hall [18] determined the natural linewidth of a particular hyperfine component of the $3p\ ^2P_{3/2}$-$3s\ ^2S_{1/2}$ transition in cooled sodium atoms in a magneto-optical trap and extracted the lifetime of the $3p\ ^2P_{3/2}$ state with an uncertainty of $\pm 0.22\%$ only.

The contradiction between measurement and theory appears to be resolved now. The new experimental results are not only in very good mutual agreement, they also agree with the results obtained from *ab initio* calculations. On account of the new results, the older BGLS measurements of Gaupp, Kuske, and Andrä [1] on sodium must be considered to be superseded now, although the quoted uncertainty of $\pm 0.16\%$ is still among the lowest ones of all measurements discussed here. However, there can be no doubt that the new BGLS measurements of this work represent a substantial improvement from the technical viewpoint (more precise alignment, lower dead times, etc.) and take into account more systematical effects in the evaluation and in the estimation of the uncertainties.

The agreement of the semiempirical calculations of Theodosiou and Curtis [6] with the measurements of Gaupp, Kuske, and Andrä [1] is thus revealed as a fortuitous coincidence. The semiempirical result of Laughlin [7], however, is closer to the new experimental values. One should keep in mind that the treatment of a complex atom as an effective one-electron problem is necessarily incomplete (for a discussion, see Brage, Fischer, and Jönsson [3]); therefore one should not expect an accuracy much better than 1% from this type of calculation, even in such favorable cases as the resonance transitions in alkali atoms.

The best agreement with the new experimental results can be stated for the multiconfiguration Hartree-Fock calculation with an approximate treatment of core-core-polarization (MCHF-CCP) calculation of Brage, Fischer, and Jönsson [3] and for the recent large-scale multiconfiguration Hartree-Fock and configuration interaction (MCHF+CI) calculation of Jönsson *et al.* [5]. The earlier all-order relativistic many body perturbation theory (RMBPT) calculation of Guet, Blundell, and Johnson [4] is also in agreement with experiment if the quoted theoretical uncertainty is combined with the experimental one. The line strength result from the coupled-cluster calculation of Salomonson and Ynnerman [11], however, shows a sizable deviation from the experimental result.

For the hyperfine constant A of the $3p\ ^2P_{3/2}$ level the situation appears less clear than for the line strengths (see Table I). Our result, the new result of Scherf *et al.* [15], and the earlier results of Krist *et al.* [12] and of Carlsson and Sturesson [13] are in good mutual agreement but disagree with the value quoted by Yei, Sieradzan, and Havey [14] with a very small uncertainty. As for the line strength the A factor resulting from the large-scale MCHF+CI calculation of Jönsson *et al.* [5] is in perfect accordance with our experimental value.

In summary, the radiative lifetimes of the first excited states in sodium were measured with uncertainties of about $\pm 0.13\%$ by means of the cascade-free technique of beam-gas-laser spectroscopy. The new experimental results from the present BGLS study as well as from other experimental sources show unambiguously that the often discussed discrepancy between *ab initio* line strength calculations and the measurements of Gaupp, Kuske, and Andrä [1] for the sodium resonance transition was caused by unrecognized systematical errors in the experiment. The present lifetime measurements confirm the most recent large-scale *ab initio* line strength calculations within experimental accuracy.

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