Accurate Lifetime Measurements of Fine-Structure States of Neutral Lithium by Beam-Gas-Laser-Spectroscopy

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Abstract

The radiative lifetimes of the $2p\ ^2P$, $3s\ ^2S$, and $3d\ ^2D$ terms of neutral lithium were measured at improved precision ($\pm 0.14\%$, $\pm 0.23\%$, and $\pm 0.1\%$, respectively) by means of beam-gas-laser-spectroscopy. We also give the line strengths for the corresponding transitions $2s$–$2p$, $2p$–$3s$, and $2p$–$3d$. For all three transitions we find an excellent agreement with recent large-scale *ab initio* calculations.

1 Introduction

The three-electron atom lithium has always been a testing ground for atomic-structure calculations [1]-[11]. Its comparatively simple structure allows for very accurate calculations of line strengths by *ab initio* methods. State-of-the-art *ab initio* calculations using methods like CI-Hylleraas expansions (Yan and Drake [9]), multi-configuration Hartree-Fock (Froese Fischer *et al.* [11]) or all-orders many-body perturbation theory (Blundell *et al.* [2]) now have the potential to determine line strengths for the lithium atom with relative uncertainties at the 10^{-4} level and better. The results obtained by the various methods are generally in very good mutual agreement.

The longstanding disagreement between *ab initio* theories and the experimental result of Gaupp *et al.* [12] for the $2p$ lifetime was resolved recently in favor of theory by a new beam-gas-laser-spectroscopy lifetime measurement performed in our group in 1995 (Volz and Schmoranzer [13]). This result, however, as well as the lifetime result for the $3s$ level of lithium reported in the same paper, were of preliminary character and left some room for improvement. In the present work we give the final results of a set of new, improved lifetime measurements on the lithium levels $2p$, $3s$, and $3d$. The uncertainties have been narrowed down to $\pm 0.14\%$, $\pm 0.23\%$, and $\pm 0.1\%$, respectively.

Due to the limited space, only a brief description of the experiment and the improvements made will be given here. A full report including more experimental details and a more comprehensive discussion will be published in the near future [14].

2 Experimental method

In beam-gas-laser-spectroscopy (see *e.g.* [15, 16]) a monoisotopic ion beam with an energy of about 160 keV is partially neutralized and unselectively excited by collisions in a gas cell. The beam then passes perpendicularly through the resonator of a linearly polarized cw dye laser in which the atomic level of interest is excited. The fluorescence photons emitted by the excited atoms are collected by a movable fiber-optics detection system, transmitted to the entrance slit of a

monochromator, and finally detected by a photomultiplier. A similar fixed fiber-optics detection system serves as a monitor to normalize the signal of the movable system. Laser- and ion-beam choppers are used together with digital lock-in counting techniques to obtain a purely laser-induced, background free and cascade free signal. A decay curve consists of 30 data points at equidistant positions of the movable detector along the beam and covers at least three lifetimes. For a lifetime measurement, about 30 decay curves are recorded and evaluated by means of a least-squares fit. Zero-field quantum beats due to unresolved hyperfine-structure are included in the fit function if needed. Finally, the lifetime (before corrections) of the excited level is obtained from the averaged individual decay constants, using the precisely measured beam velocity as conversion factor.

3 Corrections and uncertainties

The largest systematical effect in beam-gas-laser-spectroscopy arises from the divergence of the atom beam which causes a slight decrease in the total detection efficiency for the fluorescence photons as the fibre-optics detector is moved downstream along the beam. As a result the decay of the excited state fluorescence appears to be faster and the lifetime appears to be shortened by, typically, a few 0.1%. The correction procedure for this effect was a major contributor to the uncertainties in our previous measurements [13]. The procedure used there was based on an approximation of the atom beam profiles and of the spatial detection efficiency of the photon collection system by Gaussian-shaped model functions [15]. This resulted in a simple analytic expression for the beam divergence correction in which the measured parameters of the atom beam (widths, divergence angles) and of the fluorescence detector (width of the spatial detection efficiency) were inserted. In the present work the measured profiles of the atom beam and of the photon detection efficiency were fed into a computer program which calculated the beam divergence correction by numerical integration. This reduces the uncertainty in the correction by a factor of 2. The corrections applied are $(+0.62 \pm 0.07)\%$ for the $2p$ level, $(+0.94 \pm 0.10)\%$ for $3s$, and $(+0.33 \pm 0.04)\%$ for $3d$.

Another important error source in lifetime measurements is the occurence of quantum beats. The geometry of the detection system in our experiment is designed to suppress quantum beats by a magic angle geometry. Nevertheless, residual quantum beat modulations may appear in the decay curves due to the finite acceptance angle ($\pm 5^\circ$) of the detection system. A small modulation with an amplitude of $\pm 1\%$ of the total signal found on the decay curve of the $2p$ level could be assigned to the hyperfine-structure of the $2p\ ^2P_{3/2}$ state. In this case the proper theoretical expression for the quantum beat was included into the fit function. An evaluation of the decay curve without accounting for quantum beats would have yielded a lifetime about 0.3% shorter than the true value. In the case of the $3d$ level quantum beats cannot be ruled out. However, the measured decay curve displayed no significant departure from an exact exponential behavior. For the $3s$ level, finally, quantum beats are absent due to the low J of 1/2.

Other error sources included in the uncertainty estimates are the beam velocity, adjustment and linearity of the detector drive, changes in the light-guide transmission, detector saturation, quenching by residual gas, Zeeman quantum beats, and finally, statistical uncertainties. The total uncertainties of $\pm 0.14\%$ ($2p$), $\pm 0.23\%$ ($3s$) and $\pm 0.10\%$ ($3d$) result from a quadratic addition of the individual contributions and should be interpreted as single standard deviations (1σ). Compared to our previous results for the levels $2p$ and $3s$ [13], the uncertainties could be narrowed down further, in particular for the $3s$ lifetime. This reduction in uncertainty is for the most part due to the improved correction procedure for the atom beam divergence effect. In the case of the $3s$ level, increased count rates and therefore lower statistical uncertainties also contributed to the improvement.

4 Results and discussion

The results of our lifetime measurements and the corresponding line strengths are shown in Table 1 together with a selection of other experimental results with quoted uncertainties below ±1%. For the $2s$–$2p$ transition our result matches two recent more accurate results obtained by Martin *et al.* [19] and

Table 1: A selection of experimental line strengths S_{if} and upper state lifetimes τ_i for transitions in neutral lithium (uncertainties in parentheses).

Transition	**Authors(year)**	**Ref.**	**Method**	τ_i**(ns)**	S_{if}**(a.u.)**
$2s$–$2p$	Gaupp, Kuske, and Andrä (82)	[12]	BGLS	27.29(4)	32.78(5)
	Carlsson and Sturesson (89)	[17]	pulsed laser	27.22(20)	32.86(24)
	Volz and Schmoranzer (96)	[13]	BGLS	27.11(6)	33.00(7)
	McAlexander *et al.* (96)	[18]	mol. spectr.	27.102(9)	33.008(11)
	Martin *et al.* (97)	[19]	mol. spectr.	27.13(2)	32.97(2)
	this work		**BGLS**	**27.09(4)**	**33.02(5)**
$2p$–$3s$	Volz and Schmoranzer (96)	[13]	BGLS	29.72(17)	17.84(10)
	this work		**BGLS**	**29.84(7)**	**17.77(4)**
$2p$–$3d$	Schulze-Hagenest *et al.* (77)	[20]	BGLS	14.60(13)	76.9(7)
	this work		**BGLS**	**14.589(14)**	**76.98(8)**

McAlexander *et al.* [18], the latter reporting an uncertainty of only ±0.033%. Molecular spectroscopy was used in these works to determine the $2s$–$2p$ line strength from the long-range interaction potential of the molecular $A\ ^1\Sigma_u^+$ state. The earlier lifetime measurement by Gaupp *et al.* [12] differs from all the new measurements by more than four of its standard deviations and can thus be safely ruled out. For the levels $3s$ and $3d$ our new results improve the most accurate previous measurements by Volz and Schmoranzer [13] and by Schulze-Hagenest *et al.* [20] by factors of 2.5 and 9, respectively.

Among the theoretical work listed in Table 2, only two approaches, namely the quantum Monte Carlo simulation by Barnett *et al.* [8] and the Brueckner approximation by Liaw and Chiou [7], fail to agree with experiment. The other theoretical line strengths for the three transitions investigated are in excellent agreement with the experimental results which confirms the high quality level reached by *ab initio* calculations for neutral lithium.

References

[1] G. Peach, H. E. Saraph, and M. J. Seaton, *J. Phys. B* **21**, 3669 (1988)

[2] S. A. Blundell, W. R. Johnson, Z. W. Liu, and J. Sapirstein, *Phys. Rev. A* **40**, 2233 (1989)

[3] A.-M. Mårtensson-Pendrill and A. Ynnerman, *Physica Scripta* **41**, 329 (1990)

[4] A. W. Weiss, *Can. J. Chem.* **70**, 456 (1992)

[5] J. Pipin and D. M. Bishop, *Phys. Rev. A* **45**, 2736 (1992)

[6] K. T. Chung, *AIP Conf. Proc.* **274**, 381 (1993)

[7] S.-S. Liaw and F.-Y. Chiou, *Phys. Rev. A* **49**, 2435 (1994)

Table 2: Large-scale *ab initio* calculations of line strengths S_{if} for transitions in neutral lithium (pair entries L/V denote length and velocity form).

Transition	**Authors(year)**	**Ref.**	**Method**	S_{if} **(a.u.)**
$2s$–$2p$	Peach *et al.* (88)	[1]	Close coupling	33.03
	Blundell *et al.* (89)	[2]	RMBPT all orders	32.99/33.02
	Mårtensson-Pendrill *et al.* (90)	[3]	Coupled clusters	33.01
	Weiss (92)	[4]	CI	33.03/33.12
	Pipin and Bishop (92)	[5]	CI-Hylleraas	33.00
	Chung (93)	[6]	FCPC	33.00/33.00
	Liaw and Chiou (94)	[7]	Brueckner approx.	33.11/33.08
	Barnett *et al.* (95)	[8]	Monte Carlo sim.	32.83(3)
	Yan and Drake (95)	[9]	CI-Hylleraas	33.0056
	Godefroid *et al.* (96)	[10]	MCHF	33.00/33.00
	Froese Fischer *et al.* (97)	[11]	MCHF+Breit-Pauli	33.0024
$2p$–$3s$	Peach *et al.* (88)	[1]	Close coupling	17.77
	Blundell *et al.* (89)	[2]	MBPT all orders	17.76/17.75
	Chung (93)	[6]	FCPC	17.75
	Liaw and Chiou (94)	[7]	Brueckner approx.	17.89/17.89
	Froese Fischer *et al.* (97)	[11]	MCHF+Breit-Pauli	17.7505
$2p$–$3d$	Pipin and Bishop (92)	[5]	CI-Hylleraas	76.97
	Chung (93)	[6]	FCPC	77.00/77.01
	Yan and Drake (95)	[9]	CI-Hylleraas	77.0098
	Froese Fischer *et al.* (97)	[11]	MCHF+Breit-Pauli	77.0133

[8] R. N. Barnett, E. M. Johnson, and W. A. Lester Jr., *Phys. Rev. A* **51**, 2049 (1995)

[9] Z. C. Yan and G. W. F. Drake, *Phys. Rev. A* **52**, R4316 (1995)

[10] M. R. Godefroid, C. Froese-Fischer, and P. Jönsson, *Physica Scripta T* **65**, 70 (1996).

[11] C. Froese-Fischer, M. Saparov, G. Gaigalas, and M. Godefroid, *At. Data Nucl. Data Tables*, to be published

[12] A. Gaupp, P. Kuske, and H. J. Andrä, *Phys. Rev. A* **26**, 3351 (1982)

[13] U. Volz and H. Schmoranzer, *Physica Scripta T* **65**, 48 (1996)

[14] A. Schmitt, U. Volz, H. Liebel, M. Kohl, R. Henkel, H. Schmoranzer, to be published

[15] U. Volz, D. Marger, H. Roth, and H. Schmoranzer, *J. Phys. B* **28**, 579 (1995)

[16] U. Volz, M. Majerus, H. Liebel, A. Schmitt, and H. Schmoranzer, *Phys. Rev. Lett.* **76**, 2862 (1996)

[17] J. Carlsson and L. Sturesson, *Z. Phys. D* **14**, 281 (1989)

[18] W. I. McAlexander, E. R. I. Abraham, and R. G. Hulet, *Phys. Rev. A* **54**, R5 (1996)

[19] F. Martin, M. Aubert-Frécon, R. Bacis, P. Crozet, C. Linton, S. Magnier, A. J. Ross, and I. Russier, *Phys. Rev. A* **55**, 3458 (1997)

[20] D. Schulze-Hagenest, H. Harde, W. Brand, and W. Demtröder, *Z. Phys. A* **282**, 149 (1977)