Experimental lifetimes for selected laser and other levels of Ar$^+$

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Using the beam-gas method with gas-jet and doubly differentially pumped gas-cell targets, we have measured mean radiative lifetimes for several levels of Ar$^+$. The levels and lifetimes in nanoseconds are as follows: for $4p$ $^4P^o_{5/2}$, 8.28 ± 0.41; $4p$ $^4P^o_{3/2}$, 8.40 ± 0.59; $4p$ $^4P^o_{1/2}$, 8.42 ± 0.42; $4p$ $^2D^o_{5/2}$, 9.23 ± 0.46; $4p$ $^2P^o_{3/2}$, 9.19 ± 0.46; $4p'$ $^2F^o_{7/2}$, 8.47 ± 0.42; $4p'$ $^2P^o_{1/2}$, 4.50 ± 0.45; $4p'$ $^2D^o_{5/2}$, 7.84 ± 0.39. The lifetimes of the $4p$ $^2D^o_{5/2}$ and $4p$ $^2P^o_{3/2}$ laser levels are in excellent agreement (within 3%) with selective excitation measurements by Bennett, Kindlmann, Mercer, and Sunderland (1964–65).

INTRODUCTION

Radiative lifetimes are fundamental atomic and molecular parameters and as such have considerable importance. In the case of singly-charged argon, accurate radiative lifetimes are required for analysis of laser processes, in addition to being needed for checks on atomic structure calculations. They also serve as anchor points for scaling arc measurements of transition rates.

Although numerous studies have been made of the Ar II lifetimes, a considerable spread in the reported values exists. The selective excitation measurements by Bennett et al.[1] are generally considered to be the most reliable, but unfortunately they were carried out on only a few levels. Most of the other measurements were made using the beam-foil technique. One notable omission by Bennett et al. was the $4p$ $^4P^o$ term. Transitions originating from the $4p$ $^4P^o$ levels play an especially important role in wall-stabilized arc measurements.[2,3]

These considerations, we felt, justified yet another experimental investigation. We have, consequently, used the beam-gas technique to measure the lifetimes for eight levels of Ar II, including especially the $4p$ $^4P^o$ levels. Two of the levels which we investigated (the $4p$ $^2D^o_{5/2}$ and $4p$ $^2P^o_{3/2}$ laser levels) were also studied by Bennett et al. Excellent agreement between our results and their results gives us considerable confidence in the other work reported herein.

References to all known previous experimental work on Ar II can be found in a review paper by Head, Head, and Lawrence.[4]

EXPERIMENTAL TECHNIQUE

In our version of the beam-gas method, a fast ion beam (20–30 keV) is passed through either a differentially pumped gas-jet target or a doubly differentially pumped gas-cell target in order to excite the appropriate levels. The radiative decay of these levels is then observed spectroscopically downstream from the target. Because the ions are moving with a known speed v, it is easy to convert the observed exponential spatial decay of line intensity into a temporal decay from which the lifetime can be extracted. Details of the apparatus, technique, and references to our earlier work with this method can be found in Refs. 5 and 6.

ANALYSIS AND RESULTS

Decay curves are known to follow the form[7,8]

$$I = Ae^{-x/v\tau_k} + \sum_{j>k} B_j e^{-x/v\tau_j} + C ,$$

where I denotes the intensity of a particular transition originating from level k, j is a level which cascades into k, x is the distance downstream from the target, v is the speed of the ions, and A, B_j, and C are constants. C is usually included to account for background effects and for the small amount of repopulation of the levels arising from residual gas in the observation region; these are effectively constant in our experiment. Experimental points fitted by the above equation using a computer program run on a DEC PDP-10 computer. This program minimizes the least-squares sum by adjusting the parameters A, B, C, τ_k, and τ_j with standard iteration techniques. When a preset convergence criterion is met, the final values of the parameters are printed along with their standard deviations. Also computed and printed is the χ^2 integral for the fit.

Almost all of the Ar II decay curves were found to be best fitted with two exponentials and a constant. Resulting lifetimes are reported in Table I. All decay curves used passed the χ^2 integral test with lower and upper limits set at 0.03 and 0.97, respectively. Results reported are weighted means of several measurements.

TABLE I. Radiative lifetimes of selected $4p$ and $4p'$ levels of Ar^+.

Level	λ (Å)	This study[a]	BKMS[b]	Lifetimes (nsec) Recommended[c] term averages	Other[d]
$4p\,^4P^o_{5/2}$	4806	8.28± 0.17(5%)			10.3± 0.19 [12], 10.4± 2.1 [15]
				8.4 (10%)	9.5± 0.9 [13], 12.2± 2.2 [14]
$^4P^o_{3/2}$	4421	8.40± 0.24(7%)			9.6± 0.11 [12], 9.2± 0.9 [16]
					11.6± 2.1 [14]
$^4P^o_{1/2}$	4848	8.42± 0.15(5%)			11.4± 2.0 [14]
$4p\,^2D^o_{5/2}$	4880	9.23± 0.19(5%)	9.1± 0.6	9.8 (3%)	9.4± 0.6, 10.1± 0.4 [9]
$4p\,^2P^o_{3/2}$	4765	9.19± 0.14(5%)	9.4± 0.5	8.7 (5%)	
$4p'^2F^o_{7/2}$	4610	8.47± 0.14(5%)		9.1 (7%)	8.8± 0.2 [10]
$4p'^2P^o_{1/2}$	4132	4.50± 0.24(10%)		4.3 (10%)	4.0± 0.4 [11]
$4p'^2D^o_{5/2}$	4482	7.84± 0.08(5%)		7.6 (10%)	6.27± 0.12 [10], 3.69± 0.13 [10]

[a] Errors following the weighted means from several decay curves are standard deviations of the means. Our estimates of total uncertainties are given in parentheses.

[b] Reference 1. These lifetimes were obtained by selective excitation using pulsed electron beams and are considered to be the most reliable numbers available.

[c] Reference 4. These numbers represent "best estimates" for term averages based on the experimental measurements through approximately June, 1975.

[d] Selected lifetime measurements by other investigators. The number of the appropriate reference is given in brackets. References to other known previous measurements are given in Ref. 4.

One of the most serious potential errors in measurements which do not use selective excitation of the levels arises from cascading from higher shorter-lived levels to the level under investigation (the so-called growing-in cascade). A semilogarithmic plot of the data which appears to be a straight line to the eye can nonetheless contain enough cascade contribution to produce a 20% error. We have found that the only sure way to detect such cascade effects is to systematically delete points from the beginning of the decay curves and to watch the trends in the computed lifetimes and χ^2 integrals. If a growing-in cascade is present, the computed lifetimes will first be observed to drop with successive deletions until they stabilize about a certain value or begin rising as effects of long-lived cascades take over. In most cases the stabilization of the computed lifetimes is accompanied by acceptable χ^2 integrals, whereas before deletion of points no acceptable χ^2 integrals are obtained. Our standard operating procedure calls for accepting the first fit yielding an acceptable χ^2 integral in order that we not bias the results.

In addition to using printed results we also inspected computer generated plots of the data and all fits to the data, including those from the systematic deletion of points. Examples of such plots can be found in earlier work.[5-8]

DISCUSSION OF RESULTS

Inspection of the results in Table I shows that the foregoing procedure yields excellent agreement with the selective-excitation work of Bennett et al. in the case of the $4p\,^2D^o_{5/2}$ and $4p\,^2P^o_{3/2}$ levels. We also obtained agreement within statistical error with their work for the $4p\,^2D^o_{3/2}$, $4p\,^2P_{1/2}$, and $4p\,^2S^o_{1/2}$ levels, but our work was based on only one acceptable decay curve for each of those levels. We have thus not reported those results.

Two other selective excitation studies have been made on levels of Ar^+. One is a pulsed laser experiment on the $4p\,^2D^o_{5/2}$ by Arrathoon and Sealer.[9] Using two different approaches, they obtain one result which is in agreement with our work and the work of Bennett et al. and one result which is not. The other study was performed by Camhy-Val, Dumont, and co-workers.[10] Their pre-1975 work involving delayed coincidence techniques using photons in cascade appears to not be accurate; however, we find agreement with their 1975 work on the $4p'\,^2F^o_{7/2}$ level.

The $4p'\,^2P^o_{1/2}$ level has not been extensively studied. The best previous result known to us is the phase-shift measurement by Matilsky and Hesser.[11]

Our results for the $4p\,^4P^o$ levels are around 8.4 nsec, which is considerably lower than the other results included for comparison. With the excep-

tion of the delayed-coincidence work by Bakos, Szigeti, and Varga,[12] they are beam-foil studies.[13-16] Not shown in the table is a beam-foil photographic-plate measurement of 7.1 ± 2.1 by Fink, Bashkin, and Bickel,[17] which is in agreement with our values but has very large error limits.

Also included in Table I for comparison are term averages of best experimental values recommended for use at the last Beam-Foil Spectroscopy Conference.[4] Because there are sometimes large variations among lifetimes for different J values within a term, one finds some mild disagreement with those numbers.

This study indicates, at least for the levels investigated in this work, that the beam-foil results are generally about 10–20% too high. In past beam-gas work from this laboratory we have also generally allowed for as much as a 10% error due to cascade. That this has been a wise course of action is indicated by a drop of about 10% in the value of the final lifetime for the $4p\ ^2D^o_{5/2}$ level from that given in a preliminary report.[18] Interestingly, the value for the $4p\ ^2P^o_{3/2}$ level did not change. We now feel that the error brackets can now be reduced to about ±5% when statistical errors are approximately ±1%. We believe that a systematic analysis, such as we have used, of beam-foil and non-selectively-excited coincidence data would yield much more reliable results. A similar conclusion was reached by Carré et al.[19]

It may be concluded that the elimination of cascade using cascade-free techniques does not necessarily guarantee good results. We are, of course, basing much of the proof on comparisons with the work of Bennett et al. We should thus note the work of Bennett and Kindlmann[20] on Ne I using the same apparatus and technique has been independently verified by Harde and Guthöhrlein using a beam-gas-laser technique.[21] Additionally, it may be concluded that the beam-gas method, with careful attention paid to analysis, can yield results almost as reliable as those obtainable in selective excitation experiments. Since there is an enormous difference in the time and effort required by these two approaches, we feel that there is ample justification for using the beam-gas technique to provide many of the lifetimes needed in laser, theoretical, and astrophysical studies involving the first few spectra of most elements.

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