Lifetime of the $3p\ ^2P_{3/2}$ level in sodiumlike bromine (Br XXV)

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Using the beam-foil spectroscopy technique, we have measured the decay time of the $3p\ ^2P_{3/2}$ level in sodiumlike bromine. The results, using an arbitrarily normalized decay curves analysis, are in good agreement with the most recent theoretical calculations. They also confirm an earlier experimental result in sodiumlike krypton, suggesting that some previous measurements of the lifetimes of $3p\ ^2P_{3/2}$ states of nearby isoelectronic ions are too high. [S1050-2947(98)06507-X]

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INTRODUCTION

Wavelength and lifetime measurements and calculations of the resonance line transitions in the alkali-metal isoelectronic sequences are the most extensive among many-electron atoms. The case of a single s-electron ground state outside a closed np^6 shell always gives rise to a strong resonance transition to the $(n+1)\ p$ state, allowing precision spectroscopic measurements, and also accurate atomic structure calculations. For example, the $6s$-$6p$ transitions in cesium have been the focus of very precise laser-excitation measurements [1] and calculations [2] in support of observations of atomic parity nonconservation [3]. Very precise determinations of the dipole transition moment for neutral Li [4], Na [5], and K [6] have recently been made in studies of low-temperature molecular dimer spectra accomplished through laser trap technology. These transition moments can be related to the corresponding exponential lifetimes within the assumptions inherent in the Weisskopf-Wigner formulation [7], but the accuracy of these relationships has not been tested to these levels of precision in the 0.1% range [8]. The $3p\ ^2P_{1/2,3/2}$ lifetimes of the sodium isoelectronic sequence have been measured over a large range of nuclear charge up to $Z=79$ (Au68+) for the $J=1/2$ level [9], and up to $Z=54$ (Xe43+) for the $J=3/2$ level [9].

Recent theoretical developments for these alkalilike sequences have also yielded more precise transition rates: Johnson *et al.* [10] have applied the many-body perturbation theory (MBPT) to obtain accurate solutions to the wave equations in the sodium sequence. These calculations are improved over their previous calculations [2] by including all third-order random-phase approximation (RPA) corrections. Two other calculations, in particular, show good agreement with their results: the multiconfigurational Hartree-Fock calculations of Kim and Cheng [11] typically yield lifetimes 4–5 % less than those of Johnson *et al.* [10], while semiempirical calculations of Theodosiou and Curtis [12] differ by 2% or less for the whole isoelectronic sequence.

At the low-Z end of the sequence good agreement is obtained between theory and experiment, recent laser measurements in sodium itself having resolved a longstanding discrepancy [5]. However, the measurements of Hutton *et al.* [13] indicated that the $3p_{3/2}$ transition rates for ions in the intermediate range of $Z=22-29$ in Ti, Fe, Ni, and Cu are systematically about one standard deviation (5–10 %) below all the most accurate and recent theories. More recent measurements in Nb [14] and in Xe [9] also indicated the same small but systematic deviation from theory, whereas measurements of the $3p_{1/2}$ decay rates agree with the same set of calculations. Thus, there is a slight theoretical or experimental discrepancy for the $3p_{3/2}$ decay rate, at least in this intermediate Z region.

The authors of Ref. [14] have more recently applied the beam-foil decay curve technique to study the same decays in $Z=36$, Kr XXVI [15]. Their result, with a precision quoted at 5%, gives a value for the $3p_{3/2}$ rate slightly above the theoretical values. This result would suggest that the theoretical values might be correct and the earlier work of Hutton and collaborators [9,13,14] might be too low, with an error of the same order as their quoted error limits. It is clearly desirable for an independent measurement to be made for a sodiumlike ion in this range. Hence, we have made precise measurements of the $3p_{3/2}$ decay rate in Br XXV.

EXPERIMENT

We used the beam-foil technique to measure the decay lifetimes of the 189-Å resonance transition $3s\ ^2S_{1/2}-3p\ ^2P_{3/2}$ in sodiumlike bromine, Br XXV. We used a 190-MeV energy bromine-ion beam produced at the Argonne ATLAS accelerator. The 14+ charge-state beam was momentum-analyzed and directed into the foil-target chamber on the atomic physics beamline. Many experimental details of this system have been described previously [16]. For these measurements, photons emitted approximately 90° to the ion-beam axis were analyzed by the 600-l/mm grating of a McPherson 247, 2.2-m grazing incidence monochromator equipped with a microchannel-plate position-sensitive

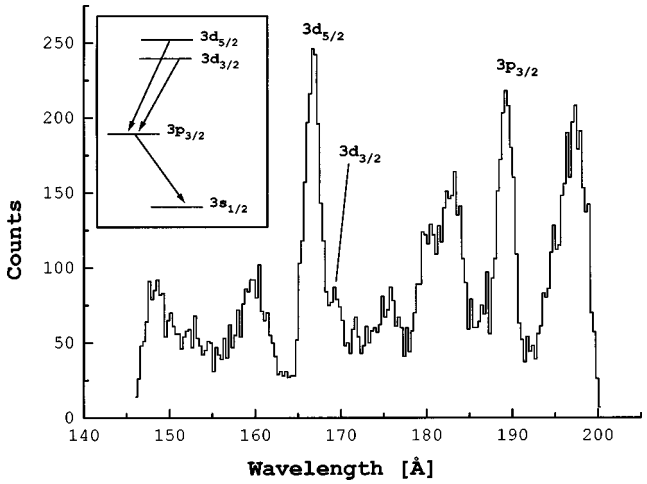

FIG. 1. A partial channel-plate spectrum taken at an observing position close to the exciter foil. The inset shows the relevant transitions used in the ANDC analysis. All transitions in the spectrum have been identified.

photon-counting detector. The standard entrance slit of the monochromator was removed, and replaced by an auxiliary slit placed on the Rowland circle, but within approximately 5 mm of the ion-beam axis. This enhanced the spatial resolution to close to the value of the slit width used, typically 0.2 mm. Spectra were normalized to beam charge collected in a shielded Faraday cup, approximately 3 m down-beam from the interaction chamber. Typical beam currents at the 6-mm-diam target were 50 nA, (3 pnA). Spectra were taken in a time-resolved mode, synchronized to the beam-pulse structure. A fast timing signal from the channel plate was used to gate the spectrum to times of arrival of the dispersed photons at the channel plate.

Decay curves were obtained by repeating data collection at 51 equidistant positions of the target foil along the beam direction. The separation between successive foil positions was 0.198 mm, corresponding to a delay time of 9.73 ps. A typical run consisted of three sets of measurements as the foil was moved up and down beam, and took several hours.

RESULTS

We show a typical spectrum in Fig. 1, with the dominant transitions identified. The $3s_{1/2}$-$3p_{1/2}$ transition occurs at 229 Å, and was observed in a separate spectrum. The beam energy of 190 MeV was selected to minimize blending from the strong magnesiumlike resonance transitions that occur at wavelengths close to sodiumlike transitions of interest here. The strongest blend is the magnesiumlike $^3P_1$ decay that occurs at the same wavelength, 167.7 Å, as the sodiumlike $3d_{5/2}$ decay. By comparison with spectra taken at lower beam energies, we verified that the blending is very small at 190-MeV beam energy. The other relevant $3s$-$3p$ and $3p$-$3d$ transitions are all well resolved from strong yrast Rydberg transitions.

We used the arbitrarily normalized decay curves (ANDC) analysis [17,18] to account for the cascading present in the $3s$-$3p$ decay curves. This cascading is dominated by the $3p$-$3d$ transitions, and hence can be easily removed using ANDC analysis applied to the $3p$ and $3d$ decay curves. The

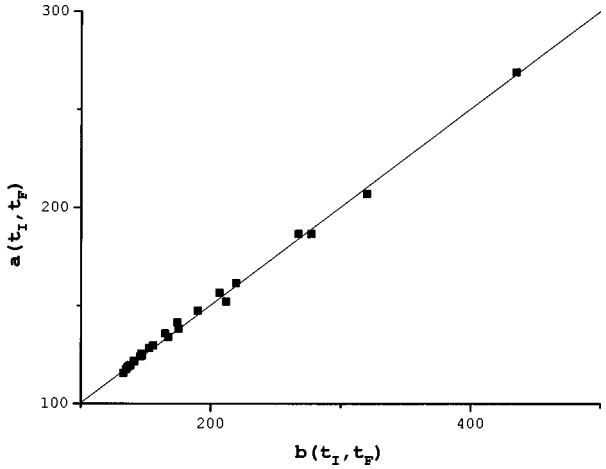

FIG. 2. An example of a single ANDC analysis. The straight-line fit yields an intercept of 50.51 ± 0.65 ps, the lifetime of the $^2P_{3/2}$ level.

two $3d$ decays into the $p_{3/2}$ state at 167 and 172 Å are partially resolved in the spectrum, and thus their decays can be treated separately, or in combination. Final results utilize the combination: both decays have the same form, since both d states ($J=3/2$ and $J=5/2$) have the same lifetimes to better than 1%, and higher n states also have lifetimes independent of fine structure to within the precision of the experiment.

We used an integral formulation of the ANDC calculation. Previous work in this system also used ANDC analyses, but with slightly different approaches [13–15]. Just as it is clear that a standard multiexponential fit to strongly cascaded decay curves may give rise to systematic errors in the lifetime results [19], it is also important to ensure that the ANDC analysis techniques used are free of systematic errors.

The ANDC equations can be written [17,18] in terms of the intensities of the primary decay curve $I_1(t)$ and its dominant cascade $I_2(t)$, which are connected through the population equation

$$\tau_1 dI_1/dt = \xi I_2(t) - I_1(t). \tag{1}$$

This was integrated over various time intervals t_I and t_F within the decay curve to yield a set of relationships

$$\tau_1 = a(t_I,t_F) - b(t_I,t_F)\xi, \tag{2}$$

where

$$a(t_I,t_F) = \int_{t_I}^{t_F} dt\ I_1(t)/[I_1(t_F) - I_1(t_I)], \tag{3}$$

$$b(t_I,t_F) = \int_{t_I}^{t_F} dt\ I_2(t)/[I_1(t_F) - I_1(t_I)]. \tag{4}$$

The quantities τ_1 and ξ were obtained by their least-squares adjustment to the experimental values of $a(t_I,t_F)$ and $b(t_I,t_F)$.

The integral form of the ANDC analysis consists of calculating $a(t_I,t_F)$ and $b(t_I,t_F)$ for several time differences to obtain a linear plot in the two parameters whose slope is the fixed relative efficiency, and with an intercept equal to the primary lifetime. An example is shown in Fig. 2 for one of our bromine runs. The $a(t_I,t_F)$ and $b(t_I,t_F)$ parameters are

TABLE I. A comparison of theoretical and experimental lifetimes (in ns) for the $3p\ ^2P_{3/2}$ level: Theoretical results are from Kim and Chen, Ref. [11] (KC), Theodosiou and Curtis, Ref. [12] (TC), Johnson *et al.* Ref. [10] (MBPT), while the experimental references are given in the last column.

Ion	Z	KC	TC	MBPT	Expt.	Ref. (year)
Si IV	14		1.132	1.140	1.14±0.05	[22] (1993)
S VI	16		0.596	0.600	0.6±0.02	[23] (1983)
Cl VII	17		0.470	0.475	0.47±0.01	[24] (1984)
Ar VIII	18	0.366	0.385	0.388	0.389±0.01	[25] (1986)
Ca X	20		0.275	0.277	0.308±0.02	[26] (1996)
Ti XII	22		0.208	0.209	0.204±0.013	[13] (1988)
Fe XVI	26	0.123	0.129	0.130	0.138±0.009	[13] (1988)
Ni XVIII	28		0.1042	0.105	0.113±0.008	[13] (1988)
Br XXV	35		0.0508	0.0512	0.0496±0.002	This work
Kr XXVI	36	0.0438	0.0458	0.0462	0.0452±0.002	[15] (1997)
Nb XXXI	41		0.0272	0.0274	0.029±0.002	[14] (1995)
Xe XLIV	54	0.006 06	0.006 27	0.006 37	0.0068±0.0006	[9] (1995)

quite strongly correlated, as pointed out by Pinnington and Gosselin [20], and discussed below, and care must be taken in deciding the choice of time intervals used. A differential form of ANDC analysis has also been used [18], the equation being written in terms of the two intensities and the logarithmic derivative at each point.

The two forms are mathematically equivalent; however, the results are differently affected by statistics of the measured decay curves. If the differential form is used with no smoothing of the input data, large variations of the ANDC plot lead to very imprecise lifetime results. The previous measurements on other members of the sodium isoelectronic sequence [13–15] have utilized the integral form; however, in addition, the original data had first been nonlinear least-squares fitted to a sum of multiexponential decays. The resulting fits were then used as inputs to an integral ANDC analysis [21]. Since the goal of ANDC analysis is to avoid the problems associated with multiple exponential analysis (which has been shown to lead to erroneous lifetimes), it is not clear whether this process is necessarily optimized or free of systematic errors.

In our analysis, we have avoided utilizing such a smoothing process. Instead, care has been used to optimize the reduction to the ANDC plot shown in Fig. 2: in a systematic study of the ANDC technique, Pinnington and Gosselin [20] pointed out that because of the strong correlations of the $a(t_I,t_F)$, $b(t_I,t_F)$, and $\Delta=[I_1(t_F)-I_1(t_I)]$ parameters, it is important that the relative errors in the three parameters be minimized and kept as equal as possible. In such cases, the standard deviations of the ANDC fit become meaningful. We verified qualitatively their suggestions: the error in each Δ parameter, which is the difference between initial and final intensities, should be minimized; this is most easily accomplished by taking time intervals, with an initial time t_I always at the peak of the decay curve, while the final time points t_F span each data set, going from the second channel to a final channel at the end of the decay. We verified that when small Δ parameters are included in the ANDC plot, such as taking short time differences on the tail of the decay curve, the poor statistics lead to curvature at each end of the plot.

The area parameters in $a(t_I,t_F)$ and $b(t_I,t_F)$ were sufficiently robust that no smoothing was necessary. Further tests included removal of data points from the front end of each data set and also from the end of each data set. In both cases, the results gave consistency within the quoted fitting error, as long as the signal-to-background ratio remained above approximately 2. Further tests included varying the number of data channels for each of the spectral lines from $3p_{3/2}$ and $3d_{3/2,5/2}$, and using the separated $3d_{3/2,5/2}$ decays. These results confirmed the stability of the results, but were not used in the final data analysis.

The average for seven sets of data leads to a lifetime for the $3p\ ^2P_{3/2}$ level of 49.5 ± 1.1 ps. Adding other errors (beam velocity 0.5%, possible systematic errors due to unknown blends, and additional weak cascades) to this value, yields a result that is accurate to 4% or 2 ps.

DISCUSSION

The goal of this work is to clarify the comparisons between theory and experiment for the $3p\ ^2P_{3/2}$ lifetime in the isoelectronic sequence for nuclear charges greater than 25.

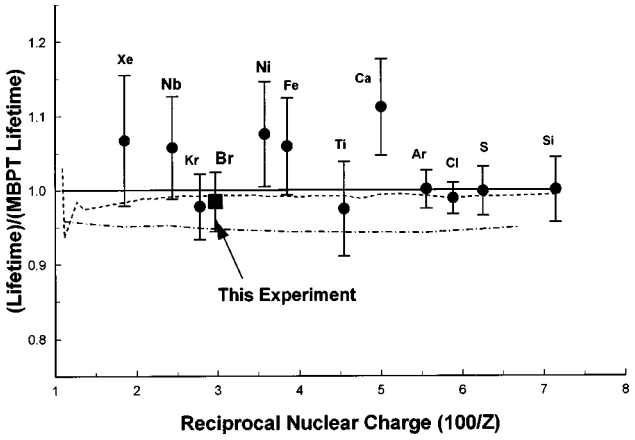

FIG. 3. Isoelectronic comparison of theory and experiment for the $3p_{3/2}$ level. The data are normalized to the many-body perturbation theory results of Johnson *et al.* [10], showing good agreement between theory and experiment for krypton [15] and bromine (this work).

Our measurements, together with those of neighboring ions are compared with theoretical values in Table I. Since the most recent many-body calculations [10] are expected to be very accurate, we have normalized all other theoretical and experimental values to their results in the graphical plot of Fig. 3 for the range $Z=14$ to $Z=54$. Our measurement agrees well with the MBPT theory, as well as with the semiempirical results [12]. Our result in Br is consistent with the result of Kink *et al.* [15] in Kr. All previous measurements had yielded lifetimes greater than theory for $Z \geqslant 25$.

Hence, our result strongly supports the conclusion of Kink *et al.* [15] that these previous experimental results may have had small systematic errors. Our analysis of the effect of statistics on the data-smoothing process in ANDC analysis suggests just such a systematic change in the predicted lifetimes, which grows as the statistics worsens. Although the work of Kink *et al.* [15] used the same data-smoothing process as those results obtained in neighboring ions, their statistics were much better, and thus, no systematic shift is seen. This data smoothing may be the reason for the previous small deviations.

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