

Laser-probing measurements and calculations of lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ metastable levels in Ba II

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The two metastable levels $5d^2D_{3/2}$ and $5d^2D_{5/2}$ in Ba II both show extremely long lifetimes of the order of several tens of seconds each. This has been found both by experiments and by theoretical predictions. The small transition probabilities associated with these two levels make them interesting and challenging for theoreticians as well as for experimentalists. Several calculations and measurements of these two lifetimes have been made previously but discrepancies between the results are present. This article presents values of $\tau = 89.4 \pm 15.6$ s for the $^2D_{3/2}$ level and $\tau = 32.0 \pm 4.6$ s for the $^2D_{5/2}$ level measured in a beam-laser experiment performed at the ion storage ring CRYRING. These values are supported by our new calculations resulting in $\tau = 82.0$ s for the $^2D_{3/2}$ level and $\tau = 31.6$ s for the $^2D_{5/2}$ level.

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I. INTRODUCTION

Singly ionized barium (Ba II) shows a very simple atomic structure: the ground state is $5s^25p^66s^2S_{1/2}$. The atomic energy levels compiled by Moore [1] and, more recently by Curry [2], belong to the $5p^6ns$ ($n=6-50$), $5p^6np$ ($n=6-12$), $5p^6nd$ ($n=5-48$), $5p^6nf$ ($n=4-14$), and $5p^6ng$ ($n=5-12$) configurations. The Ba II spectrum, and particularly the radiative lifetimes of the metastable states of this ion, are an interesting challenge for theoreticians as well as for the experimentalists. As this ion shows a rather simple electronic structure with one outer electron moving outside of closed shells in the resultant field of the nucleus and of the 54 inner electrons, outer correlation is expected to be rather small but this may not be true for core-valence correlation. In addition, as we are dealing with a rather heavy ion, relativistic effects are expected to play a role and must be considered adequately in the calculations. The lifetimes of the long-lived metastable states are traditionally an interesting test for the accuracy of the theoretical models because they are very sensitive to small configuration interaction effects.

On the experimental side a number of problems are connected with measurements of lifetimes of such long lived levels such as $5d^2D_{3/2}$ and $5d^2D_{5/2}$. Well controlled confinement of the ions is necessary and several processes such as collisional excitation and deexcitation of the levels have to be monitored to permit accurate determinations of the radiative lifetimes. Solely the ion confinement presents high technical demands on the apparatus used. In addition to an extremely good vacuum, stability over long time scales has to be obtained in order to avoid systematical errors. Prior to this study all measurements of the lifetimes of these two metastable levels in Ba II have been performed with ions in traps and in fact, to the best of our knowledge, the lifetime of the $5d^2D_{3/2}$ level presented in this study is the longest radiative lifetime ever measured in a storage ring.

A number of theoretical calculations and measurements have been published for the radiative lifetimes of the two $5d^2D_{3/2}$ and $5d^2D_{5/2}$ low-lying levels but large and unexpected discrepancies are observed among these values. For the $5d^2D_{3/2}$ level, the measurements of Schneider and Werth [3] and of Yu *et al.* [4] differ by a factor of 4 and, for the $5d^2D_{5/2}$ level, the results of Plumelle *et al.* [5] are larger than those of Nagourney *et al.* [6] and of Madej *et al.* [7] by about 50%. On the theoretical side, the scaled Thomas-Fermi-Dirac calculations of Warner [8] lead to lifetime values shorter by about a factor of 2 than the other theories both for the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ levels (Garstang [9], Guet and Johnson [10], Dzuba *et al.* [11], Gopakumar *et al.* [12], Sahoo *et al.* [13]). New measurements and calculations of lifetimes are therefore strongly needed for these two levels for definitely assessing the reliability of the different sets of published results.

II. HFR CALCULATIONS FOR FORBIDDEN LINES IN Ba II

In the present work, three different theoretical models were considered within the framework of the pseudorelativistic Hartree-Fock (HFR) method described by Cowan [14]. In a first calculation [HFR(A)], only valence correlation was considered in the physical model including the $5s^25p^6ns$ ($n=6-8$) and $5s^25p^6nd$ ($n=5-8$) configurations. In a second calculation [HFR(B)], some core-valence correlations were included by means of additional configurations with one hole in the $5s$ or $5p$ subshell. This corresponds to the following configurations: $5s^25p^6ns$ ($n=6-8$), $5s^25p^6nd$ ($n=5-8$), $5s^25p^56s6p$, $5s^25p^55d6p$, $5s^25p^54f6s$, $5s^25p^54f5d$, $5s^25p^55f6s$, $5s^25p^55d5f$, $5s^25p^54f6d$, $5s^25p^55f6d$, $5s^25p^56p6d$, $5s5p^65d^2$, $5s5p^66s^2$, and $5s5p^65d6s$. In particu-

TABLE I. Calculated A values (in s^{-1}) for $M1$ and $E2$ transitions depopulating the ${}^2D_{3/2}$ and ${}^2D_{5/2}$ metastable levels in Ba II.

Transition	Type	HFR(A)	HFR(B)	HFR(C)
${}^2D_{3/2}-{}^2S_{1/2}$	$E2$	1.357(-2)	1.217(-2)	1.219(-2)
${}^2D_{3/2}-{}^2S_{1/2}$	$M1$	0.00(0)	3.388(-13)	3.189(-13)
${}^2D_{5/2}-{}^2S_{1/2}$	$E2$	2.905(-2)	2.601(-2)	2.607(-2)
${}^2D_{5/2}-{}^2S_{3/2}$	$M1$	5.565(-3)	5.578(-3)	5.573(-3)
${}^2D_{5/2}-{}^2S_{3/2}$	$E2$	3.150(-7)	2.682(-7)	2.649(-7)

lar, it is expected that the $5s \rightarrow 5d$ core excitation plays an important role because it corresponds to an allowed quadrupole transition from the $5s^25p^65d$ configuration. The influence of $ns \rightarrow nd$ core excitation on $E2$ transition rates was already pointed out by Quinet and Hansen [15] in the case of the $3s \rightarrow 3d$ excitation in iron group elements. In our third calculation [HFR(C)], the HFR(B) model was extended with configurations characterized by two holes in the $5s$ or $5p$ subshell, i.e., $5s^25p^45d6s^2$, $5s^25p^45d^26s$, $5s^25p^45d^3$, $5p^65d^26s$, $5p^65d6s^2$, and $5p^65d^3$. Finally, as transition probabilities for magnetic dipole and electric quadrupole radiation are very sensitive to transition energies, the A values calculated using our different HFR models were corrected with wavenumbers deduced from experimental energies compiled by Curry [2]. Calculated transition probabilities for $M1$ and $E2$ transitions depopulating the ${}^2D_{3/2}$ and ${}^2D_{5/2}$ metastable levels are reported in Table I.

It is clearly seen from this table that single-electron core excitations from the $5s$ and $5p$ orbitals are much more important than double excitations. Indeed, going from calculation HFR(A) to calculation HFR(B) leads to a variation of about 10% in $E2$ transition probabilities while going from HFR(B) to HFR(C) has nearly no effect on the computed A values. Radiative lifetimes computed in the present work are compared with previous theoretical and experimental results in Tables II and III.

For the ${}^2D_{3/2}$ level, our value is in very good agreement (within 2%) with the theoretical data obtained by Guet and Johnson [10], Dzuba *et al.* [11], Gopakumar *et al.* [12], and Sahoo *et al.* [13]. For the ${}^2D_{5/2}$ level, our value is in good agreement (within 5%) with the ones obtained by Garstang [9], Dzuba *et al.* [11], and Sahoo *et al.* [13] while it is about 17% smaller than the calculations due to Guet and Johnson [10] and Gopakumar *et al.* [12]. However, these discrepancies can easily be explained by the fact that these latter authors did not take the $M1$ contribution into account. From our HFR(C) calculations, it is seen that inclusion of the $M1$ transition effectively decreases the lifetime of the ${}^2D_{5/2}$ level from 38.3 to 31.6 s. The lifetimes obtained with the most elaborated theoretical model [HFR(C)] are in very good agreement (within the error bars) with the experimental values published by Nagourney *et al.* [6], Yu *et al.* [4], Madej *et al.* [7], and those measured in the present work.

III. EXPERIMENTAL METHOD

The experiment utilized a laser probing technique (LPT) [16–18] together with a beam of Ba^+ ions stored in the ion

TABLE II. Comparison between theoretical [calculation HFR(C), see the text] and experimental lifetime (in s) for the ${}^2D_{3/2}$ metastable level in Ba II.

	Theory		Experiment	
	This work	Others	This work	Others
$5d\ {}^2D_{3/2}$	82.0	72.1 ^a 45.4 ^c 83.7 ^e 81.5 ^g 81.4 ^h 80.086(714) ⁱ	89.4(15.6)	17.5(4) ^b 48(6) ^d 79.8(4.6) ^f

^aGarstang [9].

^bSchneider and Werth [3].

^cWarner [8].

^dKnab-Bernardini *et al.* [26].

^eGuet and Johnson [10].

^fYu *et al.* [4].

^gDzuba *et al.* [11].

^hGopakumar *et al.* [12].

ⁱSahoo *et al.* [13].

storage ring CRYRING at the Manne Siegbahn Laboratory in Stockholm, Sweden. The ions were created from solid Ba placed in a Nielsen ion source equipped with an oven. The Ba^+ ions were accelerated to 40 keV and the isotope ${}^{138}Ba^+$ was selected and injected into the storage ring. The current of stored Ba^+ ions was of the order of 0.5 μA and showed an exponential decay after injection with a maximum lifetime of approximately 100 s. This loss of stored ions is primarily caused by neutralization due to collisions between the ions and the residual gas in the ring. The ion production method creates ions in a variety of different states, among them the

TABLE III. Comparison between theoretical [calculation HFR(C), see the text] and experimental lifetime (in s) for the ${}^2D_{5/2}$ metastable level in Ba II.

	Theory		Experiment	
	This work	Others	This work	Others
$5d\ {}^2D_{5/2}$	31.6	33.2 ^a 19.0 ^c 37.2 ^e 30.3 ^g 36.5 ^h 29.856(296) ⁱ	32.0(4.6)	47.0(16) ^b 32.0(5) ^d 34.5(3.5) ^f

^aGarstang [9].

^bPlumelle *et al.* [5].

^cWarner [8].

^dNagourney *et al.* [6].

^eGuet and Johnson [10].

^fMadej *et al.* [7].

^gDzuba *et al.* [11].

^hGopakumar *et al.* [12].

ⁱSahoo *et al.* [13].

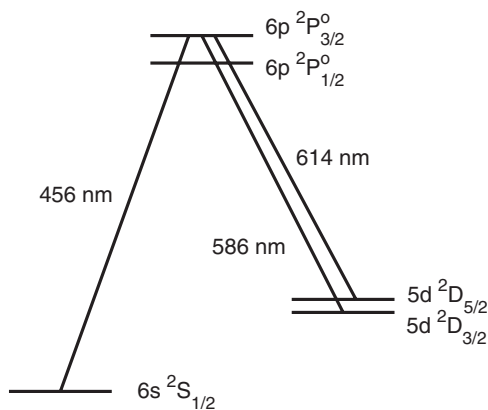


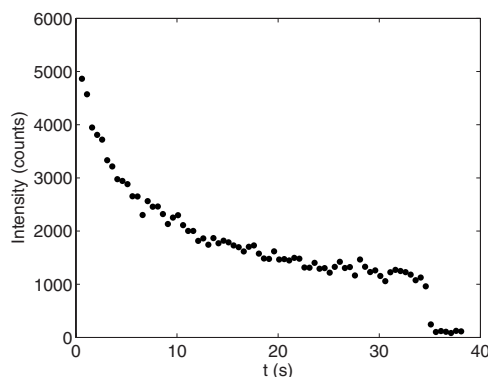
FIG. 1. Level scheme for Ba II.

metastable ones. The population of both levels ${}^2D_{3/2}$ and ${}^2D_{5/2}$ was probed through a laser induced excitation to ${}^2P_{3/2}^o$, a level that quickly decays via an $E1$ transition to the ground state ${}^2S_{1/2}$, as illustrated in Fig. 1, with a lifetime of 6.27 ± 0.25 ns [19–23]. The intensity of the laser induced fluorescence (LIF) from the transition ${}^2P_{3/2}^o$ to the ground state, which is an indirect measurement of the population of the metastable level at the time of the probe pulse, was detected by a photomultiplier tube (PMT).

A Coherent 699-29 Autoscan ring dye laser pumped with a Coherent Innova 400-25 Ar-laser was used together with the dye Rhodamine 6G which enabled lasing at both the required probing wavelengths with an output power of about 700 mW. The lifetime curves are built up by a series of data points where each data point corresponds to one cycle, i.e., one injection of ions into the storage ring. In each cycle a laser pulse induced the transition from the metastable level to the ${}^2P_{3/2}^o$ level with a time delay, relative to the ion injection, increased for each cycle. After the excitation, the fluorescence from ${}^2P_{3/2}^o$ to the ground state was detected with a Hamamatsu R585 PMT used together with a color filter that prevented the scattered laser light from reaching the detector. In this way the population of the metastable level as a function of time after injection was monitored.

In addition to the lifetime measurements the PMT was also used to measure the repopulation of the metastable level by making use of a second probe pulse that was fixed in time at the end of each cycle. Since the duration of the first probe pulse was chosen long enough (≈ 200 ms) to deplete the metastable level the second pulse measured the repopulation of the level at a time interval equal to the time difference between the two pulses. By keeping the repopulation pulse fixed at the end of each cycle this interval varied and a repopulation curve was built up [18,24] backwards.

A multichannel plate (MCP) particle detector is mounted at the end of a zero degree prolongation of one of the straight sections of the storage ring. With this detector neutralized ions which were unaffected by the magnetic field of the bending magnet in front of the MCP were detected. The stored ions get neutralized through collisions in the storage ring and the number of neutralized particles detected is proportional to the number of ions stored at the same time. The MCP was used to measure the ion beam current decay over

FIG. 2. Unnormalized fluorescence measured by probing the ${}^2D_{5/2}$ level at base pressure. Each time point represents one value of the delay time between ion injection and the probe pulse.

time and also to measure the number of injected particles in the ring at a fixed time each cycle. These two curves enabled compensation of the recorded fluorescence signals both for the decay of the ion beam and also for the possible variation in the number of particles injected during each cycle.

The number of injected particles during each cycle is, however, not necessarily directly related to the number of injected ions populating the metastable level. The ratio between these two quantities has occasionally been observed to be nonconstant during previous experiments. Indications are that the conditions under which the ion source is operated, such as gas pressure, anode voltage, and filament current, may lead to variations in the populations of different states. Therefore every fourth cycle was used to measure the LIF at a fixed time after injection. This allowed normalization with respect to variations in the initial population of the metastable level. The typical variations in the present study were about 10%.

The measured decay rate of the metastable level will consist of two parts, the radiative decay rate and a collisional decay rate. The latter is pressure dependent and by measuring the lifetime as a function of pressure the radiative decay rate can be determined by extrapolating the measured decay rates to zero pressure in a Stern-Vollmer plot [25]. A number of curves showing the decay of the LIF over time were recorded at the best possible vacuum in the storage ring, i.e., $p < 10^{-11}$ Torr. To enable the extrapolation in the Stern-Vollmer plot the pressure was later increased by heating one of the nonevaporative getter pumps connected to the storage ring. This allowed the lifetimes of the two levels to be measured at slightly different pressures. Since the pressure in the storage ring is so low, even for the high pressure measurements, no absolute measurements can be made with vacuum meters. Therefore the lifetime of the ion beam which is dependent on the number of neutralizing rest gas particles is used as an indirect relative pressure measurement.

IV. ANALYSIS

One example of the decay of the LIF is shown in Fig. 2. The curve shows the expected exponential decay and also the background signal from the PMT recorded at the end of each

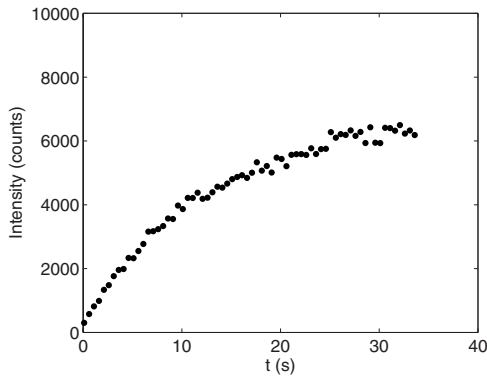


FIG. 3. Unnormalized repopulation, measured by probing the $^2D_{5/2}$ level at base pressure.

cycle after the insertion of an ion beam dump. This background was subtracted and the fluorescence curve was then normalized against the variation in original population of the metastable level. A nonzero equilibrium level can be seen before the beam dump at the end of the fluorescence curve. This is due to a continuous repopulation of the metastable level after injection into the storage ring. The repopulation of the metastable level was assumed to be mainly caused by collisions with rest gas and therefore the repopulation shown in Fig. 3 was normalized against the particle curve recorded at fixed time each cycle, Fig. 4. This effect can be corrected for by subtracting the repopulation from the fluorescence, however, before this can be done the repopulation also has to be normalized against the ion beam current decay, Fig. 5. Figure 6 shows the normalized fluorescence, the normalized repopulation and the difference between them, i.e., the final normalized lifetime curve.

Each corrected and normalized lifetime curve was fitted with a single exponential function to evaluate the lifetime of each data set. The standard deviation of the fit was used as the uncertainty of the lifetime. Each deduced lifetime and its corresponding uncertainty can be seen as one point in one of the two Stern-Vollmer plots, Figs. 7 and 8. The Stern-Vollmer plots were fitted with linear fits weighted against the uncertainty in each point which resulted in the radiative life-

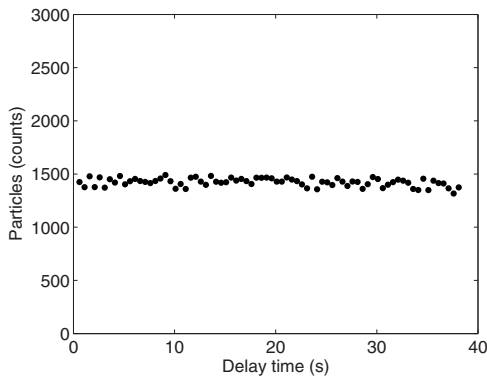


FIG. 4. Particle normalization curve recorded while measuring the lifetime of the $^2D_{5/2}$ level at high pressure. The plot shows the number of particles stored at a fixed time after injection. The time indicated on the x axis is the corresponding time point in Figs. 2, 3, and 5.

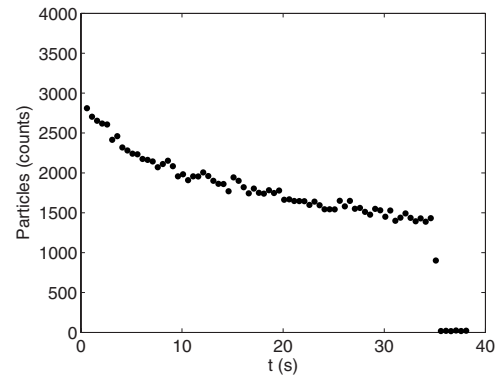


FIG. 5. The ion beam current decay at base pressure recorded while measuring the $^2D_{5/2}$ level.

time at zero pressure and a standard deviation of the fit which is listed as the uncertainty of the pure radiative lifetime.

V. RESULTS

Figures 7 and 8 show the two Stern-Vollmer plots. The weighted fit in Fig. 7 resulted in a lifetime of the $^2D_{5/2}$ level of $\tau = 32.0 \pm 4.6$ s and the fit in Fig. 8 resulted in $\tau = 89.4 \pm 15.6$ s for the $^2D_{3/2}$ level. These values are supported by our new calculated lifetimes presented in Tables II and III, $\tau = 82.0$ and 31.6 s, respectively. The results in comparison with results from previous studies are illustrated in Figs. 9 and 10.

VI. DISCUSSION

Prior to this study no measurements of the lifetime of the $^2D_{3/2}$ level showed consistent results. The first attempt to measure this lifetime was made in Ref. [3], a study in which Ba^+ ions were stored in a quadrupole trap where the lifetime was measured as a function of pressure. This allowed an extrapolation to zero pressure in order to obtain the radiative lifetime. The method used was to excite the stored ions from the ground state with a pulsed laser to the higher lying $^2P_{1/2}^o$

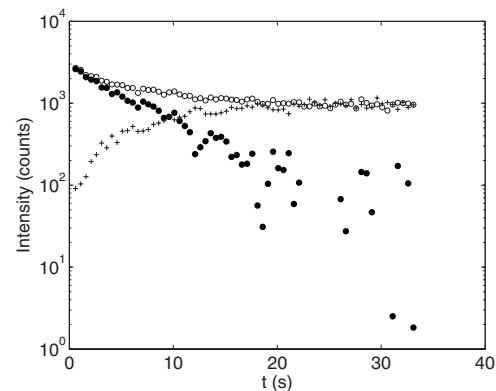


FIG. 6. Normalized fluorescence (empty circles), normalized repopulation (crosses), and the final lifetime curve (filled circles) for a measurement of $^2D_{5/2}$ at base pressure.

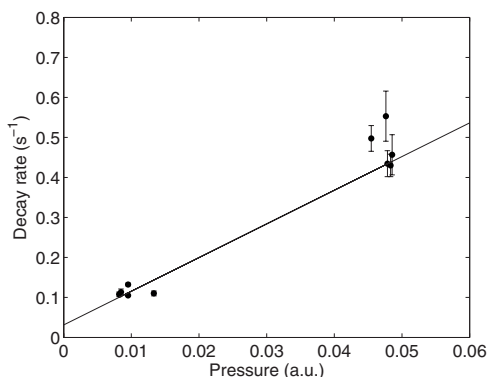


FIG. 7. Stern-Vollmer plot for the $^2D_{5/2}$ level.

level which decays partly to the ground state and partly to the metastable $^2D_{3/2}$ level with known transition probabilities. The ion cloud interacted with laser pulses sent into the trap with a repetition rate of approximately $3-30\text{ s}^{-1}$ in order to probe the ground state population which enabled the lifetime of the metastable level, or at least its lower limit, to be extracted, $\tau=17.5\pm 4\text{ s}$. The second measurement of the $^2D_{3/2}$ lifetime was made in Ref. [26]. Once again an ion trap was used but with a slightly different method to measure the deexcitation of the metastable level. The same excitation wavelength as in the first study was used but now for optical pumping into the metastable level. Once equilibrium had been reached the laser was switched off and with various delay times the ground state population was probed. The deduced lifetime of the metastable level after extrapolation to zero pressure was $49.0\pm 5.9\text{ s}$. In both these measurements one of the reasons for the discrepancies between theory and experiments was suggested to be collisional quenching due to rest gas in the ion trap. To control or monitor this process in a sufficiently accurate way is not trivial. The third measurement performed in Ref. [4] used a single barium ion stored in a trap under conditions where the authors found the collisional quenching to be insignificant. The measurement utilized the method of quantum shelving and the resulting lifetime was $\tau=79.8\pm 4.6\text{ s}$.

In the present study, however, the repopulation effect due to collisions as well as collisional deexcitation of the metastable level is of major importance as can be seen in Figs.

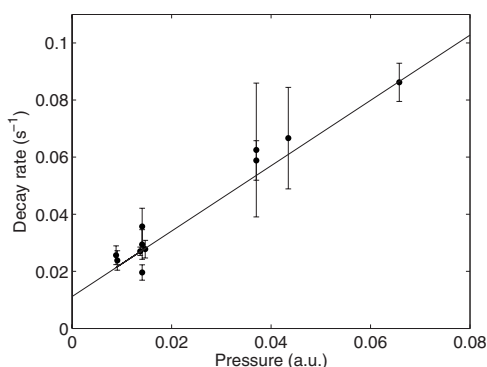


FIG. 8. Stern-Vollmer plot for the $^2D_{3/2}$ level.

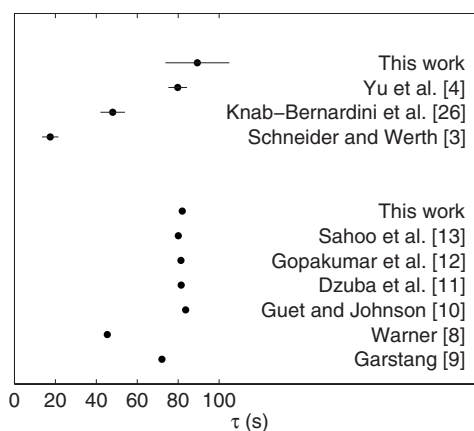


FIG. 9. Comparison between calculated and measured lifetimes for the $^2D_{3/2}$ level.

6–8. After just 20 s almost all of the fluorescence signal is originating from ions that are excited into the metastable level due to repopulation and correcting for this is of most importance. This effect has been studied several times prior to this study at CRYRING and a method for correction has been developed [18,27] and used for determining lifetimes as long as 28 s in Ti II [28]. In this particular case, changing the amount of repopulation by a factor changes the lifetime of the level approximately with twice that factor. More explicitly, if the overall intensity of the repopulation curve would be changed by 5% before subtraction from the fluorescence, the extracted lifetime would change by about 10%. Understandably this is one of the most important reasons for our fairly large uncertainties, especially in the measurement of the $\tau=89.4\pm 15.6\text{ s}$ lifetime. In order to justify the amount of subtracted signal, matching the equilibrium level of the LIF with that of the repopulation can be done if the measurement is performed under a sufficiently long time, see Fig. 6. A correct amount of repopulation subtracted from the LIF should render a curve showing a single exponential decay and therefore the accuracy of the fit of the final lifetime curve can also be used to control that the correct amount of repopulation was subtracted during the analysis. Every data set recorded during this study was carefully analyzed with

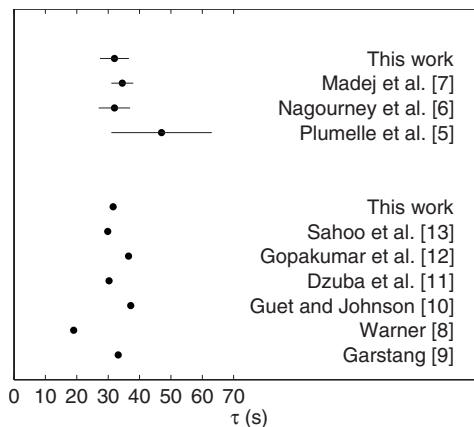


FIG. 10. Comparison between calculated and measured lifetimes for the $^2D_{5/2}$ level.

respect to repopulation and resulted in a corrected and normalized lifetime curve showing single exponential behavior.

The values presented in this article of the Ba II $^2D_{3/2}$ lifetime support the result of the previous measurement by Yu *et al.* [4], the one out of the previously existing three measurements that definitely had the most favorable experimental conditions. Thereby this study constitutes a measurement of this lifetime where a previously published result has been reproduced. The measured lifetime $\tau=89.4\pm 15.6$ s also agrees well with several theoretical values [10–13] as well as with result from our own calculation. The lifetime of the $^2D_{5/2}$ level has been measured three times prior to this study and all three measurements are within each others error bars. The lifetime presented in this article $\tau=32.0\pm 4.6$ s fits in well with these previous measurements as well as with several theoretical values [9–13] and with the calculated value presented in this article which supports not only the resulting

$^2D_{5/2}$ lifetime but also the validity of the method used for measuring also the longer $^2D_{3/2}$ lifetime.

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