

Investigation of some transitions and lifetimes in Xe II

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(Received 18 November 1993)

The lifetimes of the $6p^2 D_{5/2}^o$, $6p^4 P_{5/2}^o$, $6p^4 D_{7/2}^o$, $6p^4 D_{5/2}^o$, and $6p^2 F_{7/2}^o$ levels in Xe II have been measured with a beam-laser technique. A detailed spectroscopic investigation of the fluorescence channels of the excited states has been performed to confirm the identifications. As a consequence of some reassignments experimental lifetime values can now unambiguously be ascribed to these levels.

PACS number(s): 32.70.Fw, 32.30.Jc

I. INTRODUCTION

Many lifetime measurements of different levels in Xe II have been performed over the past years. The beam-laser technique, which was introduced by Andrä, Gaupp, and Wittmann [1], has made it possible to improve the quality of lifetime measurements by selecting a specific state without the problem of undesired cascades. For Xe II, however, there has still been some confusion concerning the lifetime of the $6p^4 D_{5/2}^o$ level for which reported results differ substantially. Mitchell *et al.* [2] recently tried to clarify the situation, however without success. Thus the origin of the discrepancy remains unresolved. We have also noted that the lifetime of the $6p^2 D_{5/2}^o$ level had not been reported as being measured by the more accurate beam-laser method, even though it is accessible by laser excitation. Recently, Hansen and Persson [3] have investigated the spectrum of Xe II. In this work a jK -coupling notation was adopted. Compared to earlier results several reassignments were introduced.

Thus we have performed an investigation of some levels in Xe II using the beam-laser method in order to clarify some of the open questions.

II. EXPERIMENT

A 25-kV electrostatic isotope separator (INIS) at the Manne Siegbahn Laboratory in Stockholm was used to produce a fast beam of singly charged Xe ions. The ions were produced in a low-voltage electron-impact ion source operated at a gas pressure of about 10^{-5} Torr. The ions were extracted by applying 25 kV and injected into a 0.5-m-radius analyzing magnet, and an isotopically pure beam was selected. For the lifetime measurements presented here the isotope ^{132}Xe was used. Two quadrupole triplets and horizontal and vertical deflection plates were used to focus and steer the beam through two 3-mm apertures 1.4 m apart. The beam was then deflected electrostatically to a Faraday cup or to an electrostatic energy analyzer, see Fig. 1. The beam current in the region between the two apertures was typically in the order of 1

μA . The experimental setup was built in UHV components, and the vacuum was in the low 10^{-8} Torr region.

A single-mode Coherent 699-29 Autoscan ring dye laser was used to provide tunable light with a bandwidth less than 1 MHz. The ring dye laser was pumped by an Innova 100-20 Ar-ion laser at 5145 Å. The laser beam was steered through a Brewster window into the vacuum system and then through the set of apertures that define the ion beam path. In this way it was possible to obtain a good overlap between the laser and the ion beam in the detection region. A beam expander was used to focus the laser beam into a chamber situated between the two apertures. It was also possible to steer the laser beam through another window to obtain excitation with the laser beam parallel instead of antiparallel to the ion beam (cf. Fig. 1). The laser frequency was controlled by a PC-version autoscan program. The dyes used were Rhodamine-6G, DCM, and Rhodamine 110.

In the chamber between the two apertures, three electrically isolated collinear tubes were mounted on a movable carriage. The carriage was connected to a precision screw for positioning inside the vacuum chamber. The two end tubes were grounded and the center tube was connected to a high voltage power supply. The ions were excited from metastable levels that were produced in the ion source. Quenching of the metastable states before the observation region could be avoided by applying high voltage to the center tube in order to fulfill the resonance criterion in the tube only. The fluorescence light was observed through a slot in the center tube. The resonance of interest was found by scanning the laser frequency for a fixed high voltage on the center tube while the fluorescence was monitored. Once the resonance was found, the overlap between the laser beam and the ion beam was optimized by monitoring the intensity of the resonance signal as the steering and focusing of the laser were varied. The acceleration voltage was calibrated by measuring the Doppler shift between excitation with the laser light parallel and antiparallel to the ion beam (see, e.g., Neugart [4]).

If the frequency of the laser light is set at resonance and the voltage on the center tube is increased, the resonance will move into the gap between the first two tubes. In this way the excitation is localized to a 1-mm region in the gap. This method was described in detail earlier by

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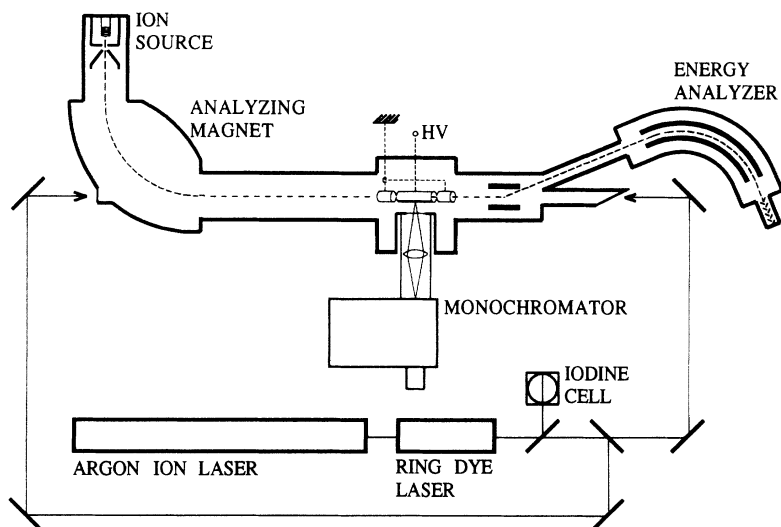


FIG. 1. Schematic diagram of the experimental setup for beam-laser measurements of atomic lifetimes.

Short *et al.* [5]. By moving the carriage it was possible to measure the variation of the fluorescence as a function of the distance from the region of excitation. A Heath EUE 700 monochromator with a cooled EMI 9789 QA photomultiplier tube was used to select and count the photons. The slits of the monochromator were set at 1 mm, which, depending on the grating chosen, gives a spectral resolution of 20–40 Å. The pulses from the PM tube were amplified and noise was removed by a discriminator. After discrimination the pulses were fed into a CAMAC counter module, and the counts were read into a PC. The PC was also used to control the step motor that moves the carriage and the voltage on the observation tube. For each position, the carriage was stopped for a certain dwell time to record the actual fluorescence intensity. The CAMAC-PC based data acquisition system has been described in more detail by Sonnek [6].

III. RESULTS AND DISCUSSION

Earlier measurements of lifetimes using the beam-foil method or the pulsed electron excitation had problems with sources of systematic errors, such as undesired cascades from higher levels. The results from such measurements were summarized by Ward *et al.* [7]. These results differ substantially from each other and have often uncertainties of more than 20%. As an example, the lifetime of the $6p^4P_{5/2}^o$ level can be mentioned. For this level experimental lifetimes from 3.2 to 22 ns have been reported [7]. The cascade-photon-coincidence (CPC) method and the beam-laser method used in this work should be free from problems associated with cascading. Mohamed, King, and Read [8] and Mitchell *et al.* [2] utilized the CPC technique, which makes use of a cascade from a more highly excited state. Photons from a transition, with the lower level as the state of interest, are collected and start a time-to-amplitude converter (TAC). The TAC is stopped by photons from the decay of this lower state. The delay between the detection of the start and stop photons is a measure of the time spent in the state of interest. The atoms are excited to all possible states by use of an electron beam. Distortion of the result due to cascades, however, is avoided by the coincidence technique.

nique.

Ward *et al.* [7] have used the beam-laser technique by which the state of interest is selectively excited by a crossed laser beam. The fluorescence from the state is measured at different positions from the excitation point in order to record the decay. Pegg *et al.* [9] have applied a technique where a collinear laser beam is used instead. In this technique the excitation point is moved through an electric field gradient by scanning the laser frequency. The movable detection systems used in the crossed beam geometry could then be replaced by a fixed one. A variation of this method has recently been developed by Jin and Church [10,11] in which the voltage is scanned while the laser frequency is kept fixed.

The earlier measurements and identifications of the spectrum of Xe II are summarized in the tables of *Atomic Energy Levels* by Moore [12], where the energies of 110 levels denoted by the *LS*-coupling scheme are given. A revised analysis of the old wavelength material was done by Hansen and Persson [3] in 1986, in which 161 energy levels were reported. The levels were noted in the *jK*-coupling scheme. This was considered to be more appropriate for Xe II since the states in general will be described by a slightly higher degree of purity in this coupling scheme [3]. Hansen and Persson also calculated lifetimes for the $6p$ levels. The levels and spectral lines discussed in the present work are shown in a partial Grotrian diagram in Fig. 2. Both *LS* and *jK*-coupling notations are given. In the present article we will mainly use the *LS* notation since this is what has been used in other similar work. This will make the discussion easier to follow.

Using cw laser light we have excited the $6p^2D_{5/2}^o$, $6p^4P_{5/2}^o$, $6p^4D_{7/2}^o$, $6p^4D_{5/2}^o$, and $6p^2F_{7/2}^o$ levels in Xe II. We have recorded both the lifetimes and the different fluorescence channels of these states.

As can be seen in Table I, there is a clear disagreement between the precision measurements of the lifetime of the $6p^4D_{5/2}^o$ state performed by Ward *et al.* [7] and Mohamed, King, and Read [8], who obtained 7.30 ± 0.21 and 8.70 ± 0.12 ns, respectively. In order to resolve this puzzling discrepancy, Mitchell *et al.* [2] recently used the

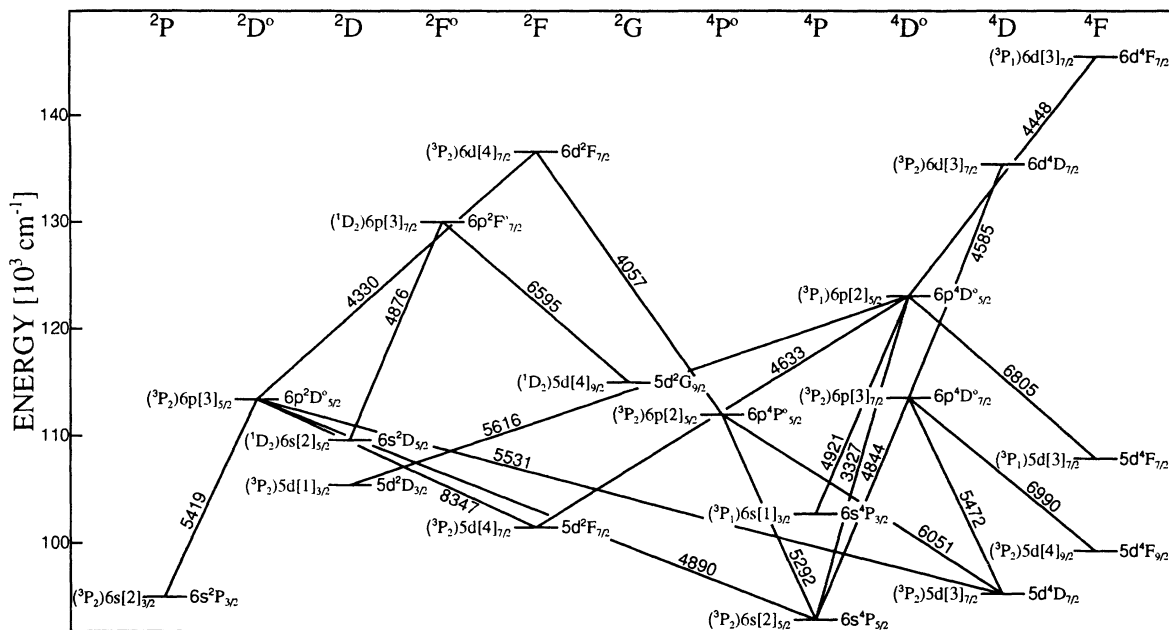


FIG. 2. Partial Grotrian diagram showing the states and transitions discussed in this article. Energies are given relative the ground state $5s^25p^5^2P_{3/2}^o$ of Xe II. As pointed out by Hansen and Persson [3] the Xe II system falls mainly in the intermediate regime and the designation of levels in jK or LS coupling is not of real significance.

CPC technique to make a new lifetime measurement. The result, 9.494 ± 0.067 ns, is significantly longer than the lifetime reported by Mohamed, King, and Read [8]. From comparison between Moore's tables [12] and the analysis of Xe II by Hansen and Persson [3] it can be seen that the assignments of several doublet and quartet levels differ. In the work by Hansen and Persson [3] the $6s^4P_{3/2}$ and the $6s^2P_{3/2}$ levels, the $6p^4D_{5/2}^o$ and the $6p^2D_{5/2}^o$ levels, and also the $6d^4F_{7/2}$ and the $6d^2F_{7/2}$ levels are interchanged compared to what was given in Moore's tables. Mitchell *et al.* [2] and Mohamed, King and Read [8] have used the $4330.52\text{-}\text{\AA}$ transition as the start channel in their measurement. This is, however, the $6d^2F_{7/2} \rightarrow 6p^2D_{5/2}^o$ transition and not the $6d^4F_{7/2} \rightarrow 6p^4D_{5/2}^o$ transition. The stop channel 5419 \AA is the transition $6p^2D_{5/2}^o \rightarrow 6s^2P_{3/2}$ and not the transition $6p^4D_{5/2}^o \rightarrow 6s^4P_{3/2}$. This indicates that the reason for

the disagreement between the measurements is that Mitchell *et al.* [2] and Mohamed, King, and Read [8] have measured the lifetime of the $6p^2D_{5/2}^o$ level instead of that of the $6p^4D_{5/2}^o$ level, which was measured by Ward *et al.* [7]. It can be noted that there is some ambiguity in the notation and classification of levels. The consequence of this discussion is that Ward *et al.* [7] measured the lifetime of a level different from that studied by Mitchell *et al.* [2] and Mohamed, King, and Read [8]. To check the new assignment by Hansen and Persson [3] we have recorded the fluorescence channels from the state excited by laser light at 5531 \AA . According to Hansen and Persson the $5531\text{-}\text{\AA}$ line should be the $5d^4D_{7/2} \rightarrow 6p^2D_{5/2}^o$ transition. In agreement with their assignments we found fluorescence at 4890 , 5419 , 5719 , and 8347 \AA (see Fig. 3). The strongest transition at 5419 \AA was used for the lifetime measurements. A lifetime of

TABLE I. Lifetimes (ns) of some energy levels in Xe II.

		$6p^2D_{5/2}^o$	$6p^4P_{5/2}^o$	$6p^4D_{7/2}^o$	$6p^4D_{5/2}^o$	$6p^2F_{7/2}^o$
Experiment	Method ^a					
This work	BL	9.3 ± 0.2	7.8 ± 0.2	6.8 ± 0.2	7.2 ± 0.2	7.3 ± 0.2
Short <i>et al.</i> [5]	BL		8.03 ± 0.10			
Pegg <i>et al.</i> [9]	BL		7.95 ± 0.16			
Mitchell <i>et al.</i> [2]	CPC	9.494 ± 0.067^b			$(9.494 \pm 0.067)^b$	
Ward <i>et al.</i> [7]	BL		7.90 ± 0.15	6.89 ± 0.20	7.30 ± 0.21	
Mohamed <i>et al.</i> [8]	CPC	8.70 ± 0.12^b	7.47 ± 0.27		$(8.70 \pm 0.12)^b$	
Mohamed and King [13]	CPC			5.82 ± 0.2		
Theory						
Hansen and Persson [3]		7.01	5.57	4.97	5.20	5.56
Garpman and Spector [14]		9.3	8.9	6.8	6.9	10.8

^aBL: Beam-laser method; CPC: Cascade-photon-coincidence method.

^bMitchell *et al.* [2] and Mohamed *et al.* [8] have probably measured the lifetime of the $6p^2D_{5/2}^o$ level instead of that of the $6p^4D_{5/2}^o$ level.

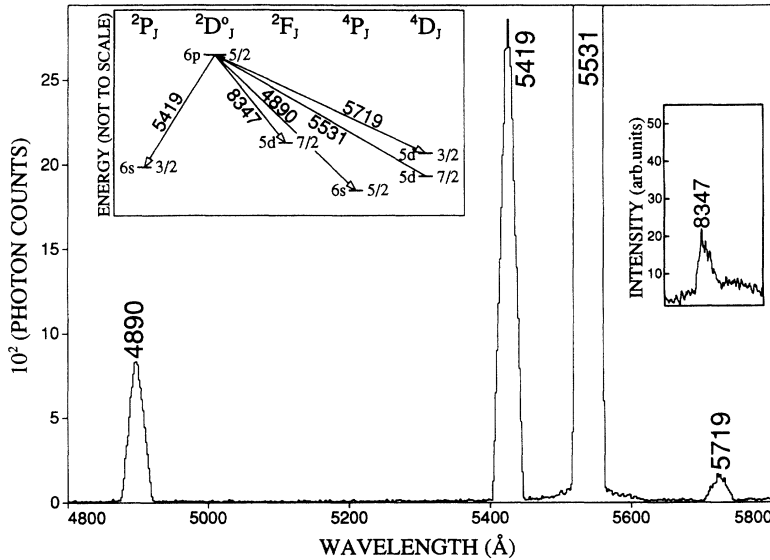


FIG. 3. Fluorescence spectrum from the $6p\ ^2D_{5/2}$ level in Xe II excited by laser light at 5531 Å. The different decay channels at 4890, 5419, 5719, and 8347 Å are shown together with the laser light at 5531 Å.

9.3 ± 0.2 ns was obtained, which is in agreement with the corresponding value given by Mitchell *et al.* [2]. Possible explanations for the lower value given by Mohamed, King, and Read [8] were discussed by Mitchell *et al.* [2]. For our measurements we conclude that the longer lifetime obtained by Mitchell *et al.* [2] is the lifetime of the $6p\ ^2D_{5/2}$ level. Ward *et al.* [7] have not performed any measurements on this level. They mention however, in their paper that the transition corresponding to 4921 Å is a fluorescence channel from $6p\ ^2D_{5/2}$, which is not correct according to Hansen and Persson [3], and also according to our measurements as can be seen below.

Mohamed, King, and Read [8] have also measured the lifetime of $6p\ ^4P_{3/2}$ level, using the 4057-Å line as the start channel which, according to Hansen and Persson [3], is a transition from the $6d\ ^2F_{7/2}$ level and not the $6d\ ^4F_{7/2}$ level. The stop channel was the 5292-Å line. The measured value of the lifetime is again somewhat lower than those of other measurements. Pegg *et al.* [9] and Ward *et al.* [7] have also measured the lifetime of this state obtaining consistent results. To check the new setup and to compare with our earlier results [5], we have remeasured this lifetime by exciting the level with laser light at 6051 Å, and monitoring the fluorescence at 5292 Å. The result in the present work, 7.8 ± 0.2 ns, is consistent with the previous reported values. In the new setup it is so far not possible to use beam energies higher than 25 keV, which is seven times lower than the energy used in our earlier work [5]. This limits the spatial resolution along the beam and thus the accuracy in the work.

The only previous measurement of the lifetime of the $6p\ ^4D_{5/2}$ level is actually that of Ward *et al.* [7], according to our reinterpretation above. Ward *et al.* [7] excited the level with laser light at 6805.74 Å. There is, however, also some confusion here. According to their paper, the dye Rh 560 was used to obtain the mentioned wavelength. But the dye necessary for obtaining the desired wavelength is DCM and not Rh 560. Furthermore, they claim that fluorescence at 5419 Å was observed, but according to the discussion above, this wavelength is emitted as the $6p\ ^2D_{5/2}$ decay. Our measurements confirm

the work of Hansen and Persson [3]. For excitation at 6805 Å we observe fluorescence at 3327, 3564, 3612, 4633, 4921, and 5616 Å (see Fig. 4). We obtained a lifetime 7.2 ± 0.2 ns which is in agreement with the value given by Ward *et al.* [7], i.e., 7.30 ± 0.21 ns. Thus, we assume that they performed measurements on the correct level and that the resolution of their fluorescence detection permitted observation of the decay at 4921 Å. In footnote 57 in the paper of Hansen and Persson [3] the problem with the measurement performed by Ward *et al.* [7] is discussed. It is, however, incorrectly pointed out that the $6p\ ^4D_{5/2}$ level corresponds to $(^3P_2)6p\ [3]_{5/2}$. According to Table III of that paper and also to our mea-

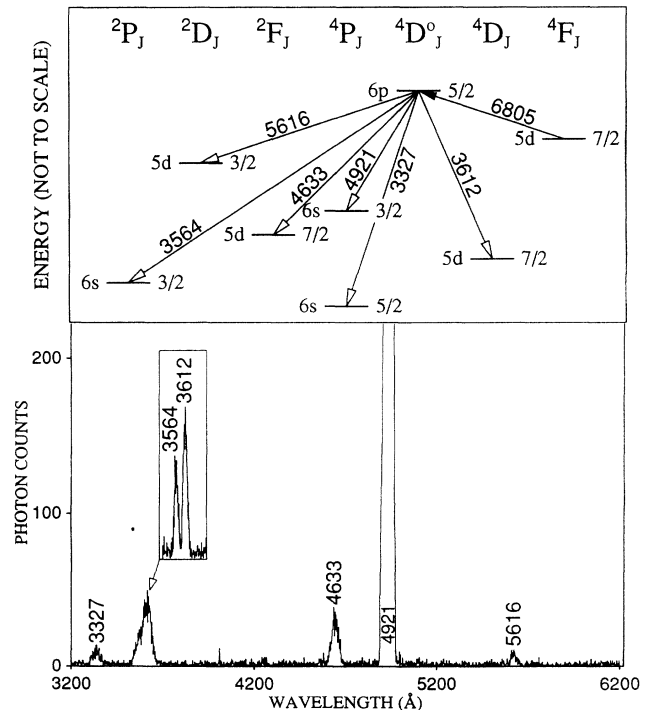


FIG. 4. Fluorescence spectrum from the $6p\ ^4D_{5/2}$ level in Xe II excited by laser light at 6805 Å. The different decay channels at 3327, 3564, 3612, 4633, 4921, and 5616 Å are shown.

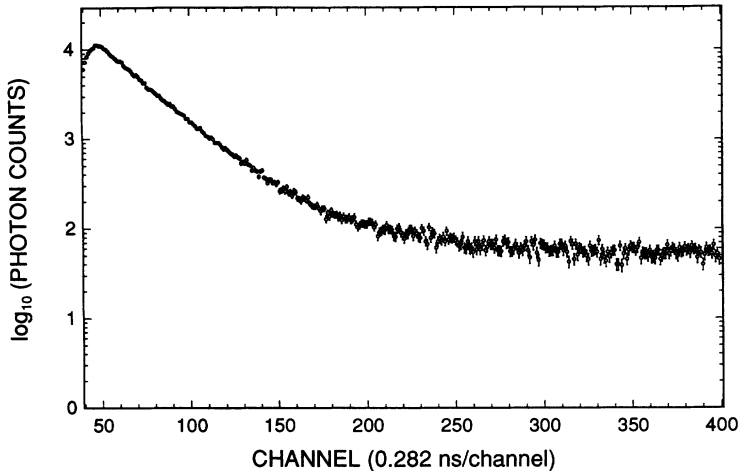


FIG. 5. Decay curve for the $6p\ ^4D_{7/2}^o$ level in Xe II. The fluorescence light at $4844\ \text{\AA}$ is displayed as a function of distance from the point where laser light at $5472\ \text{\AA}$ excited the ion beam. The measured lifetime was $6.8 \pm 0.2\ \text{ns}$.

measurements the $6p\ ^4D_{5/2}^o$ level corresponds to $(^3P_1)6p\ [2]_{5/2}$.

Mohamed and King [13] and Ward *et al.* [7] have measured the lifetime of the $6p\ ^4D_{7/2}^o$ level. Here there seems to be no controversies regarding the assignments. Mohamed and King [13] used the line $4585\ \text{\AA}$ as a start channel and $6990\ \text{\AA}$ as a stop channel. Ward *et al.* [7] have excited the level with the $5472\text{-}\text{\AA}$ transition and recorded the fluorescence at $4844\ \text{\AA}$. They obtained a lifetime of $6.89 \pm 0.20\ \text{ns}$. In the present work, we have also excited the $6p\ ^4D_{7/2}^o$ level by the $5472\text{-}\text{\AA}$ transition. In accordance to the spectroscopes work by Hansen and Persson [3] we observed fluorescence at 6990 and $4844\ \text{\AA}$. We obtained a lifetime of $6.8 \pm 0.2\ \text{ns}$ (Fig. 5) which agrees within error bars with the value given by Ward *et al.* [7]. Mohamed and King [13] found the lifetime to be $5.82 \pm 0.2\ \text{ns}$, which again is significantly lower than the values obtained in the laser experiments.

In this work, we have also measured the lifetime of the $6p\ ^2F_{7/2}^o$ level, which has never been measured with a high precision technique before. We obtained a value of $7.3 \pm 0.2\ \text{ns}$. The level was excited from the $5d\ ^2G_{9/2}$ level by laser light of $6595\ \text{\AA}$, and the fluorescence at $4876\ \text{\AA}$ was observed from the decay to the $6s\ ^2D_{5/2}$ level. The lifetime of the $6p\ ^2F_{7/2}^o$ level was discussed by Hansen and Persson [3]. They mention that the calculated values by Garpman and Spector [14] can only be regarded as an

upper limit for the lifetime values, since only the $6p \rightarrow 6s$ transitions are considered. Hansen and Persson [3] pointed out that the difference between the calculations is and should be especially large for the $6p\ ^2F_{7/2}^o$ level since the strong decay channel to the $5d$ configuration is omitted by Garpman and Spector [14]. This is in agreement with our measurements, since we obtained a lifetime which is $3\ \text{ns}$ shorter than the value of Garpman and Spector [14]. The lifetime calculations of all $6p$ levels by Hansen and Persson [3] are systematically $2\ \text{ns}$ shorter than the experimental values, while the theoretical work by Garpman and Spector [14] seems to agree better, except for the $6p\ ^2F_{7/2}^o$ level.

IV. CONCLUSIONS

Lifetime measurements have been discussed in the article by Hansen and Persson [3]. According to that work, more experimental data are needed before any firm conclusions concerning the lifetimes can be drawn. With the reinterpretation of the old lifetime measurements and with the new results from our work, the experimental values are now well established with high accuracy. We note, however, that theoretical work has, so far, not been able to deliver values in close agreement with experiment.

ACKNOWLEDGMENT

This work was supported by the Swedish Natural Science Research Council (NFR).

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