# Lifetime measurements and absolute oscillator strengths for single ionized thorium (ThII).

Physica Scripta. Vol. 38, 370–373, 1988.

# Lifetime Measurements and Absolute Oscillator Strengths for Single Ionized Thorium (ThII).

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*Received February 12, 1988; accepted May 5, 1988*

**Abstract**

Using collinear fast beam cw dye laser modulation spectroscopy accurate lifetimes in ThII have been measured. Together with known branching ratios these lifetimes have allowed the determination of absolute oscillator strengths for ThII. This element is of astrophysical interest as a chronometer for stellar and galaxy evolution, with oscillator strengths playing an important role in synthesis of stellar spectral data.

## 1. Introduction

The heavy elements are only present in stellar spectra in small amounts. Their presence is nevertheless of interest to astrophysicists, as they offer insight in stellar and galactic evolution and its connection to the timescale for nucleosynthesis [1]. The weakly radioactive nucleus $^{232}$Th is one such heavy element and to enable accurate modeling of the stellar spectral data, knowledge of absolute oscillator strengths is essential as well as a knowledge of the population mechanisms of excited levels as function of plasma temperature. A combination of branching ratio data, accurately determined lifetimes and relative intensities measured in classical emission spectroscopy can answer these questions.

Reliable lifetime data are available for many elements, both neutrals and their ionized species, using various laser-based methods. The most successful methods so far are based on the cathodic sputtering source [2], and time resolved laser spectroscopy on fast ions [3] which both have enabled precise lifetimes to be obtained in the iron-group and rare-earth elements using selective laser excitation. Only few reliable lifetime measurements however exist for the actinide elements [4, 5]. Collinear fast beam cw dye laser modulation spectroscopy is well suited for lifetime studies in complex elements as is demonstrated in this work in singly ionized Thorium. The measured lifetimes are subsequently converted to absolute oscillator strengths using branching ratios from classical light sources [6, 7, 8].

## 2. Experimental

The rich and dense optical spectra of the actinide elements are well suited for fast beam laser spectroscopy [4]. Many metastable levels are populated in the hot plasma of the ion source, enabling a wide range of levels to be excited using cw dye lasers. Furthermore the low pressure conditions in the laser–ion interaction region allow decays to be observed without any collisional deexcitation. In this particular experiment a 300 kV isotope separator, equipped with a universal ion source was used to produce $Th^+$-ions. The ions were obtained from ThO by the $CCl_4$ method and typical mass separated currents of 0.1 μA were obtained after electrostatic acceleration to 100 keV. The $Th^+$ ions were passed through a 1.2 meter long drift tube, where they were excited by the output from a frequency stabilized cw dye ring laser in a collinear geometry. In this geometry the laser excitation frequency $\sigma_L$ is given by

$$\sigma_{ion} = \sigma_L \gamma(\beta)(1 - \beta) \quad (1)$$

where $\beta = v/c$ is the reduced velocity of the ions, $\sigma_{ion}$ the ion rest frame transition frequency and $\gamma = 1/\sqrt{(1 - \beta)}$ is the time dilation factor. The laser frequency is measured using a travelling Michelson wavemeter [9].

The collinear geometry offers several advantages in spectroscopic studies of complex elements. Most importantly, the kinematic velocity compression [10] reduces the Doppler broadening, allowing for an effective excitation with narrow-band tunable dye lasers. This geometry furthermore allows a precise velocity tuning as seen from eq. (1) just by applying a postacceleration voltage to the drift tube. This postacceleration defines the interaction volume as the ions only are Doppler shifted into resonance in the postacceleration section. The resulting laser induced fluorescence was detected with a spectrometer, equipped with a photomultiplier. To reduce the noise, due to scattered laser light, the fluorescence was, whenever possible, detected on branches different from the excitation channel.

To measure excited state lifetimes, the laser was tuned into resonance and the spectrometer adjusted to select a strong decay channel. Then the laser field was pulsed, using an acousto-optic modulator. Pulses as short as 20 ns FWHM could be obtained, limited only by the finite sound velocity in the modulator. Acousto-optic modulation of a cw dye laser is advantageous, as it provides high flexibility for optimizing pulse length and pulses repetition rate for each particular decay time. Especially in fast beam work, with its low target densities, an effective duty cycle is essential. Typical laser powers were 200 mW/mm$^2$. To extract lifetimes, the laser induced fluorescence started a time to pulse height converter, while the laser pulse itself provided the stop condition. With the convolution procedure, described in the following section, lifetimes as short as 10 ns could be determined, while the upper lifetime limit was set by the flight time of app. 4 μs in the collinear interaction volume.

## 3. Results

Several decay curves were obtained for each level studied. As the laser pulse width is not negligible compared to the decay curves, the experimental data were fitted to a convolution of the exponential decay with the excitation pulse, assumed to

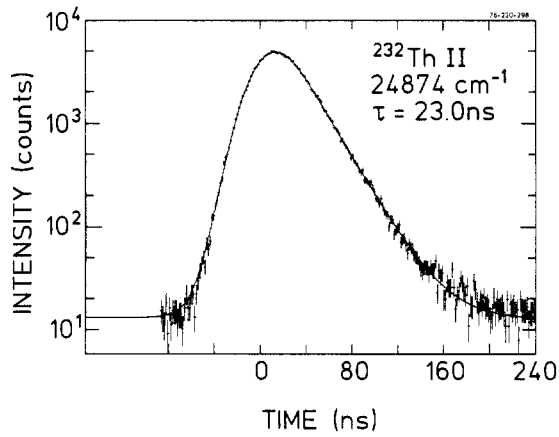


*Fig. 1.* Decay of the 24 874 cm$^{-1}$ level. A lifetime of 23.0 ± 0.6 ns was found by adjusting eq. (2) to the experimental decay curve.

be gaussian distributed. The convolution integral $g$*Exp is given by

$$g^*\text{Exp} = C\left[1 + \text{erf}\left(\frac{t-m}{\sigma\sqrt{2}} - \frac{\sigma}{\tau\sqrt{2}}\right) \times \exp\left(\frac{\sigma^2}{2\tau^2} - \frac{t-m}{\tau}\right)\right] \quad (2)$$

where the laser pulse has FWHM $\sigma$, the studied level has a lifetime $\tau$ and where $m$ is the prompt signal, determining the arrival time of the laser pulse. erf is the error function. A typical decay curve is shown in Fig. 1 together with the fitted function $g$*Exp.

A total of 18 excited levels were investigated in this work. Previously only one level was studied. Using beam-foil spectroscopy, the lifetime of the 24874 cm$^{-1}$ level was found to be 21 ± 3 ns [5] in good agreement with this work. The results are given in Table I together with the single beam-foil result. Also given in Table I are the uncertainties, composed of statistical and systematic errors. The statistical errors are negligible in all decay curves obtained, whereas the systmatic errors due to deviations from a clean collinear geometry increases with the decay time or flight time in our apparatus. The quoted uncertainties result from varying the excitation geometry around collinear geometry as well as changing the focusing of the laser and ion beams.

Table I. *Measured and calculated lifetimes in $^{232}$ThII, odd levels*

| Upper level (cm$^{-1}$) | $\tau_{exp}$ (ns)[a] | $\tau_{Corliss}$ (ns)[b] | $\tau_{LA}$ (ns)[c] |
|---|---|---|---|
| 18 214.4255 | 376 (C) | | 2420 |
| 19 050.8286 | 579 (C) | | 3300 |
| 19 248.2702 | 453 (C) | | 2280 |
| 20 686.1465 | 502 (C) | | 2220 |
| 21 131.7988 | 1290 (D) | | 3530 |
| 21 297.4160 | 315 (C) | | 1620 |
| 21 682.7467 | 807 (D) | | 3000 |
| 22 642.1052 | 3560 (D) | | 7180 |
| 24 132.0347 | 159 (B) | | 635 |
| 24 414.6410 | 62.6 (A) | 70 | 258 |
| 24 463.7894 | 94.2 (A) | 101 | 367 |
| 24 873.9828 | 23.0 (A); 21 ± 3 [5] | 18 | 123 |
| 25 188.1205 | 280 (C) | | 826 |
| 25 440.2307 | 66.2 (A) | 73 | 209 |
| 26 424.4705 | 151 (B) | | 375 |
| 27 249.5445 | 125 (B) | | 157 |
| 28 720.3154 | 15.3 (A) | 15 | 34.7 |
| 29 095.4640 | 29.0 (A) | 30 | 81.9 |

[a] (A) ±3%, (B) ±5%, (C) ±10%, (D) ±25%.
[b] Corrected, using eq. (6) and branching ratios from Corliss & Bozman, Ref. [6].
[c] Uncertainty in $\tau_{LA}$ is due to the intensity, $\Delta(I) = 8\%$.

## 4. Analysis and discussion

Lifetimes are derived from oscillator strengths, but the conversion of experimentally determined lifetimes to oscillator strengths is difficult, as it requires knowledge of branching ratios. Branching ratios are traditionally derived from spectroscopic emission data. Assuming the gas to be in local thermodynamical equilibrium (LTE) absolute intensities directly relates to oscillator strengths.

LTE implies a Boltzmann distribution

$$N_0\, e^{-E_u/kT} \cdot g_u/g_0 \quad (3)$$

where $N_u(N_0)$ is the number of atoms in the upper (lower) level. $g_u(g_0)$ is the statistical weight of the upper (lower) level, $E_u$ is the energy of the upper level, $k$ is the Boltzmann constant and $T$ is the excitation temperature in the discharge.

The emission intensity is directly related to the $gf$ value of the upper level by

$$I \propto gf\sigma^3\, e^{-E_u/kT} \quad (4)$$

where $\sigma$ is the wavenumber for the transition $u \rightarrow 0$.

A proper evaluation of oscillator strengths is thus critically dependent on absolute intensities $I$ as well as LTE being realized. Most emission sources are however not in LTE and to proper account for such deviations directly measured lifetimes can be compared to lifetimes derived from emission spectroscopy by the following relations

$$\log_{10}\left(\frac{\tau_{emission}}{\tau_{direct}}\right) = C_1 + C_2 \cdot E_u \quad (5a)$$

or

$$\log_{10}(gf) = \log_{10}(I\lambda^3) - C_1' - C_2 E_u \quad (5b)$$

where $C_1(C_1')$ and $C_2$ are constants and $E_u$ the energy of the upper level. $C_1(C_1')$ absorb errors in the absolute determination of intensities and $C_2$ determines the deviation of the temperature $T$ from a Boltzmann distribution by introducing an energy dependent correction $\Delta T$. Equation (5) assumes the relative intensities, that is branching ratios, to be correctly determined.

For ThII two sets of emission derived intensities are available, the ones of Corliss [6] and the more recent comprehensive spectra obtained at Los Alamos [8].

*A: Cu-arc spectra* [6]

Based on the Cu-arc derived ThII intensities [6], Corliss deduced oscillator strengths for a large number of ThII levels [7]. This analysis depended entirely on the single Beam-foil lifetime determination [5] of the 24874 cm$^{-1}$ level, assumed to have a branching ratio of 93% for the 4019 Å branch. Deviations from local thermodynamical equilibrium (LTE) were compensated for by assuming a ThII population similar to that of FeI in the same Cu-arc. These oscillator strengths are derived from a single line. Based on our lifetime measurements the conversion from branching ratios and lifetimes to

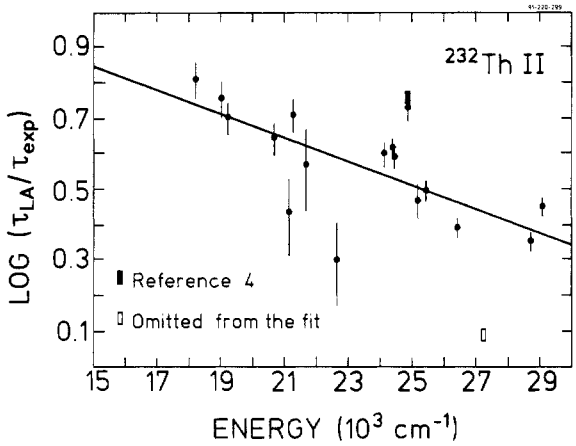


*Fig. 2.* The deviations from LTE in the Los Alamos hollow cathode are shown. The ratio log $(\tau_{LA}/\tau_{exp})$ represents the correction on the oscillator strengths assuming correct branching ratios.

absolute oscillator strengths can be put on a firmer basis using many levels distributed over a wide range of excitation energies. Using only those levels, where all major branching ratios are measured [6, 8] we can find a universal correction to the oscillator strengths of Corliss [7]. The following relation is found.

$$\log [gf] = \log [I^{NBS} \cdot \lambda^3] - 16.7414 + 8.88 \times 10^3 \cdot E \quad (6)$$

where $I^{NBS}$ are the intensities given in Ref. [6], $\lambda$ is the wavelength [Å] of the transition with $f$-value $gf$ and exited level energy $E$. Equation (6) shows, that the oscillator strengths of Corliss [7] have to be increased 25% to agree with our precise lifetimes. The deviations from LTE, given by the last term in eq. (6) is found to be in agreement with Corliss [7]. The lifetime for 6 levels, calculated with eq. (6) is shown in Table I.

*B: The Hollow-cathode spectra* [8]

A severe limitation in this analysis is not only erroneous branching ratios but branches not included at all due to incomplete identification of spectral lines. The Los Alamos atlas of the Thorium spectrum [8] presents a much more complete list of lines as well as photoelectric recorded line intensities. Assuming their hollow cathode lamp to be in LTE, they deduce temperatures for neutral and singly ionized Thorium using known $gf$ values from Corliss & Bozman [11], believed to be accurate to a factor of 2 at best. They find

$$gf = \frac{8.68 \times 10^{-15} I \lambda^2}{e^{-3.24 \times 10^{-4} E}} \quad (7)$$

for ThII oscillator strengths. $I$ is the recorded intensity of transition with wavelength $\lambda$ and upper energy level $E$. The temperature was 4450 ± 50 K. By identifying all decay branches from a given upper level, lifetimes can be determined. They are given in Table I and Fig. 2 shows the ratio $\log_{10} (\tau_{LA}/\tau_{exp})$ versus $E$, the energy of the upper level with lifetime $\tau_{exp}$. A correction to the Los Alamos $gf$ values is given by eq. (5a) assuming exact branching ratios. Except for a few levels we have good agreement with the logarithmic dependence. The relative large deviation for the 27 250 $cm^{-1}$ level must be due to incorrect assignments of lines in the Thorium Atlas [8]. In the least squares fit to the experimental data we therefore excluded this point. The levels 21 132 and 22 642 $cm^{-1}$ fall below the fitted line, also indicating wrong assignments. Our measured lifetime for the 24 874 $cm^{-1}$ level was found to be in good agreement with the previously measured Beam-foil value. To explain the deviation found for the lifetime, deduced from the Los Alamos Atlas, the branching ratio of 93% for the 4019 Å transition must be reduced to 85%. This corresponds to a 25% reduction in recorded intensity of this branch. Our final result is a correction, given by eq. (5a), with $C_1 = 1.35$ and $C_2 = -3.34 \times 10^{-5}$. These constants allow us to deduce lifetimes (oscillator strengths) from the Los Alamos Atlas with an uncertainty better than 25%.

## 5. Conclusion

Collinear fast-beam cw dye laser modulation spectroscopy has been used to measure excited state lifetimes in single ionized thorium (ThII). A total of 18 lifetimes have been measured allowing us to deduce absolute oscillator strengths for this ion using branching ratios from classical spectroscopic sources. The oscillator strengths derived by Corliss [7] and based on a single beam-foil lifetime [5] are corrected, resulting in an increase of 25%. The Hollow-cathode [8] spectra, obtained at Los Alamos, are corrected to allow a direct evaluation of oscillator strengths. Our analysis brings these two spectroscopic works in agreement and allows us to give ThII oscillator strengths to within 25%, with the uncertainty given by the incomplete knowledge of branching ratios and absolute intensities.

These oscillator strengths are of importance in the synthesis of solar and stellar spectra which in turn provide useful information on stellar evolution.

Th oscillator strength determined by Andersen and Petkov [5] and Corliss [7] for the 4019 Å ThII line (log $gf = -0.20$) corresponds to a solar Th abundance of log $\varepsilon_{Th} = 0.20 \pm 0.10$ (on a scale where log $\varepsilon_H = 12$). If the oscillator strength is increased by 25% (log $gf = -0.10$), as shown in this paper, the new solar Th abundance will be log $\varepsilon_{Th} = 0.10 \pm 0.10$. This is still in reasonable agreement with the Th abundance in carbonaceous chondrites [12] log $\varepsilon_{Th} = 0.18 - 0.23$. In particular, the new result shows that the 4019 Å ThII line can not be significantly contaminated by lines from other elements, (contamination would mean that the Th abundance derived from the solar spectrum should be *higher* than the meteoritic abundance). It is thus confirmed that the 4019 Å line gives a fairly clean measurement of the Th abundance in stars, despite the possibility of contamination from weak CoI lines [13].

## Acknowledgement

Discussions with Poul Erik Nissen is acknowledged as is support from the Carlsberg Foundation and the Danish National Science Research Council.

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