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Spatio-Temporally Resolved Dual Laser Plasma Photoabsorption Studies of Thorium Plasmas with an Extreme UV Continuum Light Source

TMR Large-Scale Facilities Access Programme

J T Costello et al

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Spatio-Temporally Resolved Dual Laser Plasma Photoabsorption Studies of Thorium Plasmas with an Extreme UV Continuum Light Source

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SUMMARY

This report describes the experiment entitled ‘Spatio-Temporally Resolved Dual Laser Plasma Photoabsorption Studies of Thorium Plasmas with an Extreme UV Continuum Light Source’; carried out at the Central Laser Facility (CLF) from the 28th July to the 25th August 1997. The experiment, funded by the Framework IV Large-Scale Facilities Access Scheme, was proposed by Dr J T Costelo, Centre for Laser Plasma Research, and carried out by visiting researchers from the Centre. They were supported by researchers from Queens University of Belfast and the Central Laser Facility, Rutherford Appleton Laboratory.

Experimental Results

- A Dual DLP experiment was performed on a 1 micron laser generated thorium plasma using the XUV continuum form the picosecond x-ray source

- Initial syudies measure the duration of the XUV from various targets to be 100-200ps for various targets (Sm, W, Au and Pb)

- Studies of the duration of the XUV continuum show that it did not depend on ASE

- Time resolved measurements clearly show that absorption was due to predominantly five times ionised Th ions, matching computed models indicating the presence of both ground state and metastable state absorption.

The CLF makes beam time at its facilities available to European Researchers with funding from DG-XII, CEC under the Large-Scale Facilities Access Scheme. For further information contact Dr. Chris Edwards at the CLF. Tel: (0)1235 445582, e-mail: c.b.edwards@rl.ac.uk

The EU researchers with collaborators from Queens University, Belfast
and RAL support staff.

From left to right:

Top row: A MacPhee, JT Costello, S Huntingdon, O Meighan, N Takeyasu

Bottom row: L Dardis, W Shaik

Arising Publications

Refereed Publications

J T Costello, O Meighan, L Dardis, C Moloney, CMcGuinness, C L S Lewis, R O'Rourke and A MacPhee, I C E Turcu, C Danson, S Huntingdon, N Takeyasu and W Shaikh. "First observation of controlled collapse of a 5f wave in the actinides: the case of thorium and its ions." In preparation to **PRL**

J T Costello, O Meighan, L Dardis, C Moloney, CMcGuinness, C L S Lewis, R O'Rourke and A MacPhee, I C E Turcu, C Danson, S Huntingdon, N Takeyasu and W Shaikh. "A 150 psec.table top laser plasma based extreme-UV continuum light source: characteristics and application to a dual laser plasma (DLP) photoabsorption experiment: " In preparation to **J.Phys.B**

Conference Proceedings

ECAMP, Sienna, Italy, July 1998.

J T Costello, O Meighan, L Dardis, C Moloney, CMcGuinness, C L S Lewis, R O'Rourke and A MacPhee, I C E Turcu, C Danson, S Huntingdon, N Takeyasu and W Shaikh "Controlled collapse of the 5f wavefunction of Thorium along its isonuclear sequence: first observation dual laser plasma photoabsorption experiments".

J T Costello, O Meighan, L Dardis, C Moloney, CMcGuinness, C L S Lewis, R O'Rourke and A MacPhee, I C E Turcu, C Danson, S Huntingdon, N Takeyasu and W Shaikh "A table top dual laser plasma based XUV photoabsorption experiment with 150 psec. time resolution; performance characteristics and results"

EQEC98, Glasgow, Scotland, Sept.1998.

O Meighan, L Dardis, C Moloney, CMcGuinness, J T Costello, C L S Lewis, R O'Rourke and A MacPhee, I C E Turcu, C Danson, S Huntingdon, N Takeyasu and W Shaikh. "Characteristics of an extreme-UV continuum light source based on a laser plasma driven by a 7psec. 248 nm pulse".

O Meighan, L Dardis, C Moloney, CMcGuinness, J T Costello, C L S Lewis, R O'Rourke and A MacPhee, I C E Turcu, C Danson, S Huntingdon, N Takeyasu and W Shaikh. "Table top picosecond dual laser plasma photoabsorption experiment"

Internal Reports

J T Costello, C McGuinness, L Dardis, C Moloney, O Meighan, C L S Lewis, R O'Rourke, A MacPhee., I C E Turcu, C Danson, S Huntingdon, N Takeyasu, W Shaikh. "Spatio-Temporally Resolved Dual Laser Plasma Photoabsorption Studies of Thorium Plasmas with an Extreme UV Continuum Light Source". RAL Annual Report 1997/98.

Spatio-Temporally Resolved Dual Laser Plasma Photoabsorption Studies of Thorium Plasmas with an Extreme UV Continuum Light Source

INTRODUCTION

When an extreme-ultraviolet (EUV) photon interacts with an atom or ion in a ground or excited state, in addition to valence electron excitation and/or ionization, several competing processes can take place involving inner shell and/or multiple electron excitations. Interpretation of the multi-electron dynamics of the excitation and decay processes requires theoretical models capable of handling the complex many-particle nature of the problem. The challenge to experimentalists is to devise techniques which can identify and measure accurately the various excitation and decay processes for atoms and ions in well described initial and final states. In addition to its fundamental interest, the study of photoexcitation/ ionization in this energy regime is also of considerable practical interest as the results are relevant to an understanding of many laboratory and astrophysical plasma processes. Furthermore, as atoms in molecules or condensed matter often exist in ionic form, measurements on their free atom or ion counterparts can prove helpful in elucidating the effects of the molecular or condensed matter host. Most recently, the discovery that nanoclusters can be formed in laser matter interactions [1)] has refocussed interest on free atom/ion spectra for comparative purposes.

For neutral atoms experimental techniques for photoionization are well developed and spectroscopies such as photoabsorption, fluorescence, photoion, photoelectron and coincidence measurements often combined with angular resolution have provided very detailed data on atomic excitation and decay dynamics [2)]. For ions the situation is much less satisfactory mainly due to the great difficulty in generating high density ion beams. An extremely versatile technique for the recording of photoabsorption spectra of atoms and ions is the Dual Laser Plasma (DLP) method [3)]. One laser plasma constitutes the 'sample' while the other becomes the XUV backlighting source. The DLP method has been used to study a wide variety of atomic and ionic species [4)] and has contributed to a number of significant advances in the field of atomic inner shell excitations over recent years [5)].

A key element of the DLP technique is the extensive 'line free' XUV continuum emission from laser plasmas of the rare-earth and neighbouring metals [6) which provides a compact and economic alternative to a bending magnet or insertion device at a synchrotron radiation facility. In a previous experiment at RAL [7) we showed that one could produce similar emission using 7ps., 248 nm pulses with an upper limit of 750 ps. on the duration. Since the time resolution of the DLP method is determined by the XUV pulse width one major goal of the current experiment was to better measure this parameter for some typical target plasmas (Sm, W, Au & Pb) and study its dependence on certain laser characteristics, in particular laser to ASE pulse energy ratio.

One of the most interesting entities to have been revealed via inner shell excitation is the so called 'Giant Resonance'. It is a characteristic of the collective nature of electron interactions in atoms and observed for $\Delta n = 0$ transitions in medium and high-Z atoms. Since first seen in the XUV spectra of the lanthanide solids in the mid 1960's they have received extensive theoretical and experimental study in atomic, molecular and solid state phases [8)]. Only two experiments on ions have been published [9,10)] in the intervening thirty years or so and a half dozen or so on the actinides (ref. 11 and refs. within). To date no experiment on photoionization of actinide ions has been published and in order to address this gap we decided to look at 5d-photoionization along the this nuclear sequence.

MATERIALS and METHODS

1. XUV Continuum Duration

A schematic of the experimental setup used to measure the XUV continuum pulse duration is shown in figure 1(a). 7psec. pulses from the frequency mixed output of a synchronously pumped dye laser were amplified up to ~ 20 mJ by double passing them through a KrF excimer laser. The KrF amplifier was triggered by a 12 Hz clock derived from the 82MHz oscillator of the modelocked Nd-YAG laser. The output from the KrF amplifier was focused to an irradiance of ~ 10^{13} W/cm^2 *in vacuo* (10^{-5} Torr) onto rod targets via a f/17.5 fused silica plano-convex lens with a focal length of 35 cm. An optical low pass filter consisting of a pair of Ag coated mirrors oriented at a grazing angle of 10° combined with an XUV transmission

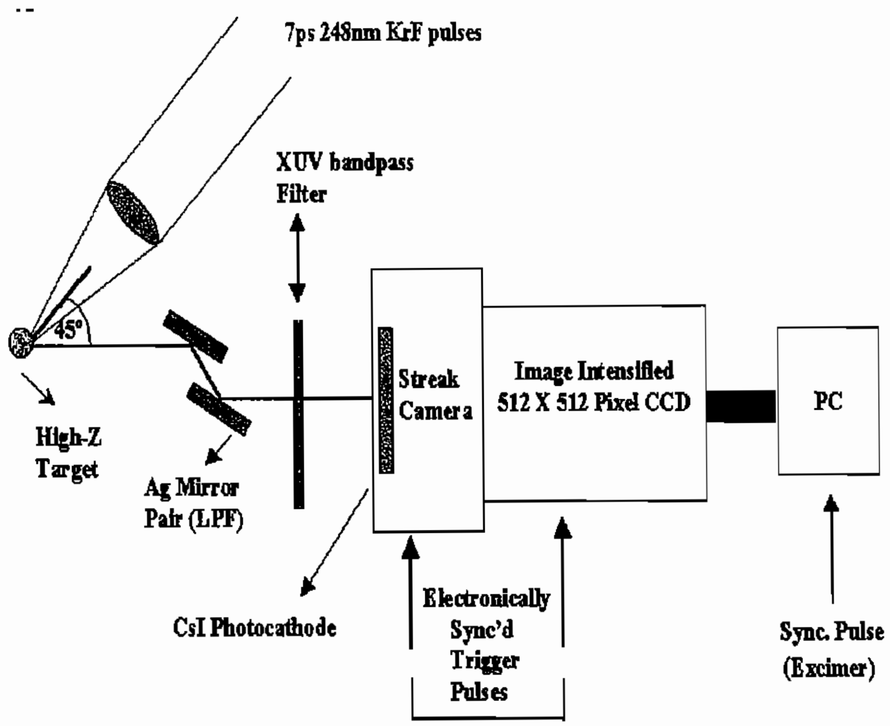

Figure 1. (a) Experimental Setup for extreme-UV continuum pulse duration measurements.

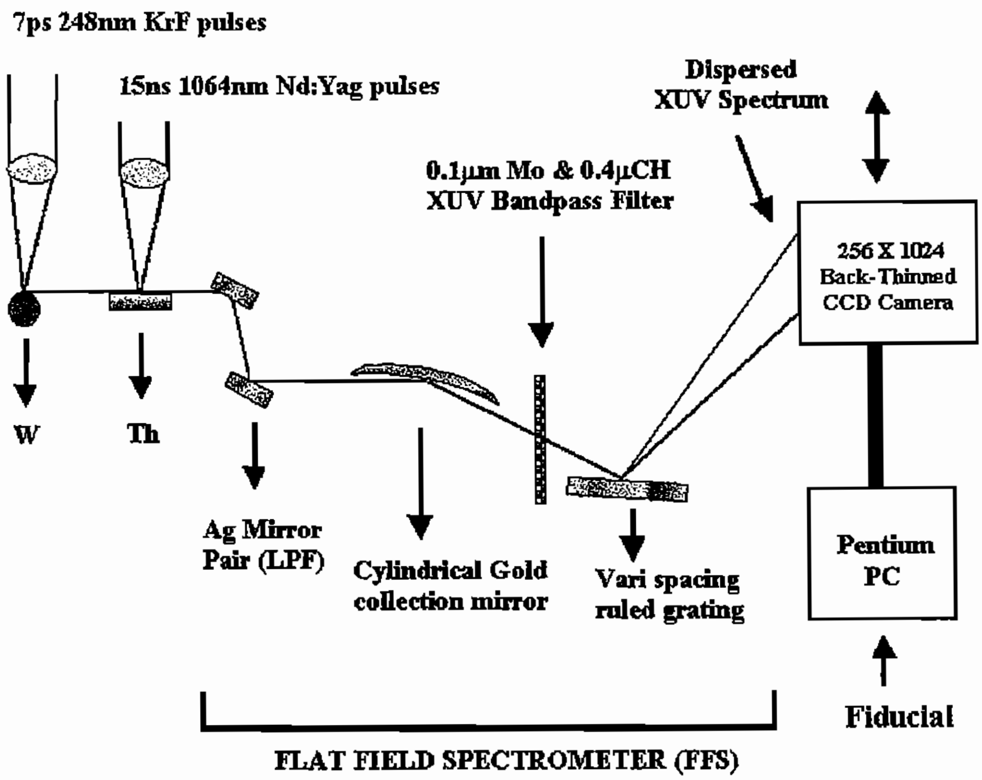

Figure 1. (b) Experimental setup for time and space resolved dual laser plasma (DLP) photoabsorption measurements.

filter composed of thin films of Mo (0.185 μm), CH (0.159 μm) and the 0.1 μm formvar substrate of the streak camera CsI photocathode yielded an symmetric transmission curve with a peak transmission of 65% @ 115 eV and FWHM of ~ 75eV. The streaked image was read out by an image intensified CCD camera and the overall system time response was better than 60 psec.

2. *Time Resolved Dual Laser Plasma Experiments*

The setup used to record time integrated emission and time resolved photoabsorption spectra is shown in figure 1 (b). The thorium absorbing plasma was generated by focussing the 1064 nm/ 300 mJ/ 8 nsec. output pulse from a Q-switched Nd-YAG laser to a spot size of ~100 μm dia. via a f/5 lens (f = 12.5 cm). The 248nm/ 20 mJ/ 7psec. pulse was synchronised to the Pockels cell of the Nd-YAG laser and, tightly focused onto a tunsten rod target, generated a backlighting XUV continuum with a variable delay and an inter laser pulse jitter of +/- 500 psec. An XUV prefilter, similar to the one used in the streak camera measurements virtually eliminated all photons above 200 eV thereby ensuring that the window of interest from 100 - 200 eV was free from order sorting problems. In addition the pre filter eliminated all scattered

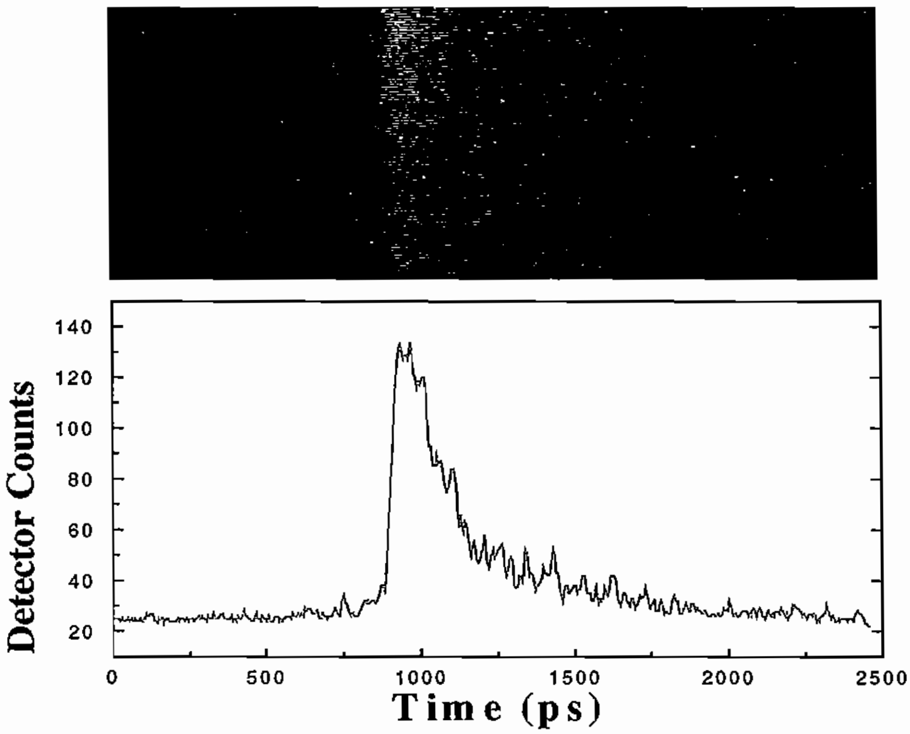

Figure 2. 2D and corresponding vertically binned CCD image of the streak from a tunsten plasma.

light permitting the measurement of reliable relative cross-sections. Transmitted XUV radiation was dispersed by a flat field spectrometer equipped with an ANDOR back thinned CCD which typically covered the 65 - 200 eV range with its 1024 pixel width. The spectral resolution achieved approached 250. The thorium target was affixed to a three axis mount which permitted us to probe different regions of the expanding plasma plume. The footprint of the XUV beam at the thorium target position was estimated to be < 100 μm V X 100 μm H as determined by the geometric acceptance of the spectrometer and its associated optics.

RESULTS and DISCUSSION

Comprehensive sets of measurements of both static and dynamic images were made for four target materials (Sm, W, Au & Pb). The static images were used to determine estimates of the streak camera instrument function for each set of data which is reasonably well approximated by a Gaussian curve of FWHM 60 psec. or so. The results are summarised in table 1. and show the mean continuum pulse duration to be sub 200 psec. or so and as low as 120 psec. for a tungsten plasma. These values may be reduced by 10 - 15 psec. after deconvolution of the streak camera response. At the irradiances used here one would expect plasma temperatures of 150 eV or so and ion velocities of ~10^7 cm/sec. Hence continuum emission times of 100 - 200 psec. correspond to plasma emitting sizes of 10 - 20 μm or so.

Continuum Target	**Average FWHM (ps)**	**Std Deviation (ps)**
Lead	123	±42
Tungsten	176	±42
Samarium	183	±27
Gold	209	±34

Table 1.

It has been well established that the duration of X-ray emission from ultrashort pulse laser plasmas tends to increase in the presence of either ASE or controlled pre-pulses [12)]. In order to investigate the effect of a 20 ns. ASE pedestal from the KrF amplifier on the continuum duration a spatial filter with an adjustable iris was used to vary the ratio of psec. laser to ASE energy (see figure 1.). We found little dependence of XUV continuum duration on ASE level for ASE energy = 0.5 -> 2.0 times picosecond laser pulse energy. Since even at the lowest ASE level we could form a pre-plasma and given that the pre-plasma parameters did not vary much over the range of ASE energy available this observation is not too surprising. Unfortunately we could not eliminate the ASE to determine the the spectral purity and duration of continuum produced in a direct laser-solid target interaction. However, one would expect to observe a further pulse width reduction in this case.

We observed a marked dependence on picosecond laser pulse energy (figure 3.). In case of tungsten, the continuum duration ranged from < 100 psec. to almost 200 psec. for pulse energies of 10 to 23 mJ or so. The growth of continuum duration with laser pulse energy appears to show a threshold at ~ 15 mJ and is very marked at the higher energy limit of the laser. This latter point explains the error of +/- 25% f.s.r. on the measurements given in table 1. since shot to shot fluctuations of up to +/-50% or so in the psec. laser energy were not uncommon.

The time evolution of XUV photoabsorption by laser produced thorium plasmas is shown in figure 4. The energy range chosen straddles the 5d-subshell ionization limits of thorium permitting the study of both 5d photoexcitation and photoionization. At long times (> 400 nsec.) after the initiation of the thorium plasma the absorption spectrum exhibits two well defined features, a narrow one below the 5d ionizations limits and a broad and strong asymmetric one above the limits. There is no single simple interpretation of the spectrum. One approach which relies on the collective response of the atom's electron cloud to an external electromagnetic field [13)] yields good agreement with the current photoelectric data and an earlier measurement using the long pulse DLP technique [14)]. There is however a discrepancy in the width of each feature which was postulated to be due to low lying metastable states in thorium and in particular of the $6d^3 7s$ configuration.

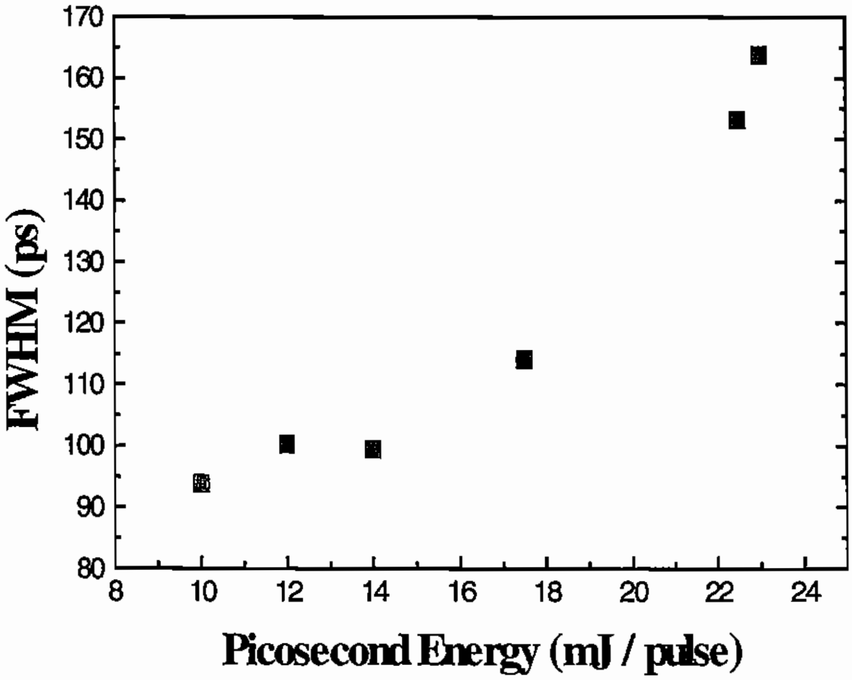

Figure 3. Dependence of extreme-UV continuum pulse duration on picosecond laser pulse energy.

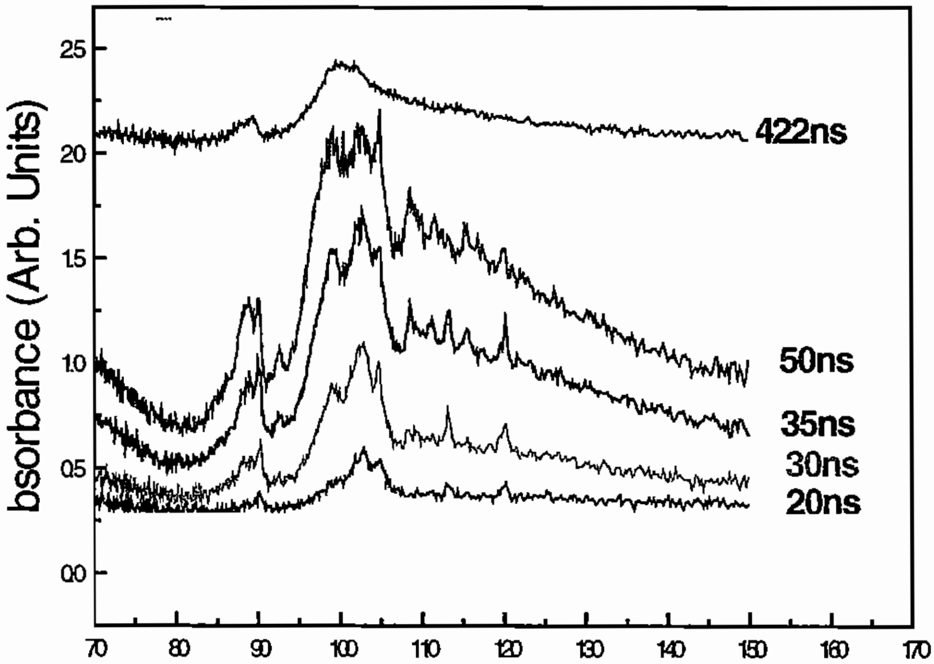

Figure 4. Time resolved extreme-UV photoabsorption spectra of a thorium plasma showing the transition from 'Giant Resonance' continuum absorption to discrete Rydberg like spectra with decreasing time delay.

As the inter-plasma time delay is shortened it can be seen that the character of the spectrum changes radically as the broad giant resonance becomes overlaid with discrete structure and Rydberg series develop. Such dramatic changes in oscillator strength distribution along an isonuclear sequence were observed only once before observed in 1981 for 4d - ε,nf excitations in the Ba /Ba^{+} /Ba^{2+} isonuclear sequence using resonant laser excitation and ionization of Ba vapour[9)]. Only very recently was this study extended to triply ionized La[10)] . The data shown in figure 4. represent the first measurements along an actinide isonuclear sequence. A simple picture requires an alternative interpretation of 'giant resonances'.

f-electrons possess bi-well potentials separated by a potential barrier. In atomic thorium a 5f (^{1}P 'like') state lies above the 5d limits but trapped by the potential barrier while the 5f (^{3}P, ^{3}D 'like') states lie below the 5d limits and are located in the inner well of the potential. The higher nf states all lie in the outer well eliminating the possibility of 5d - nf (n>5) transitions. Thus the atomic spectrum of Th may be interpreted as comprising a 5d - 5f (^{3}P, ^{3}D) low energy resonance and a 5d - 5f (^{1}P) high energy 'giant resonance'. As the degree of ionisation

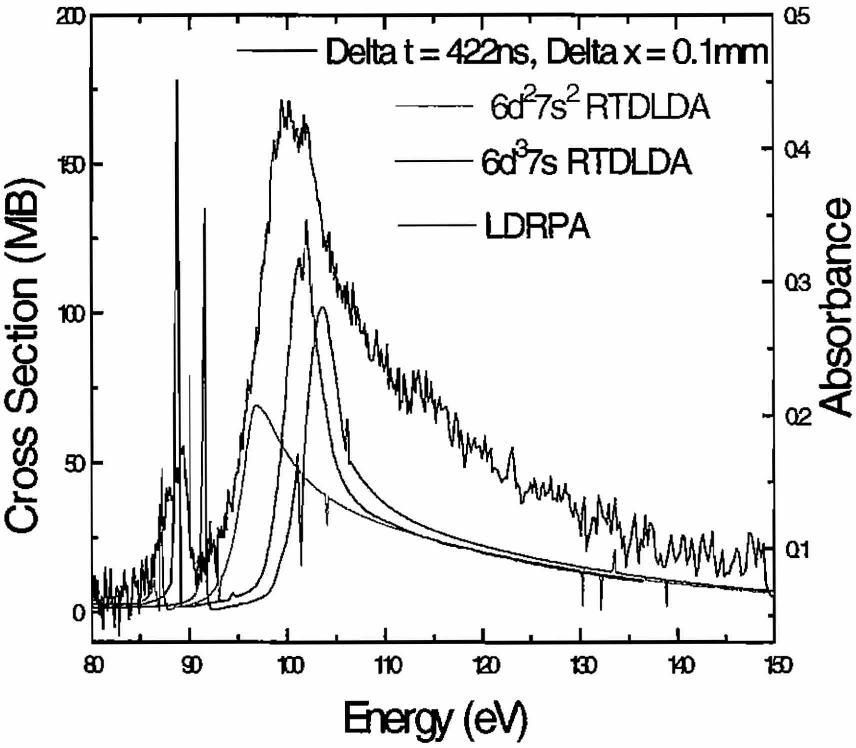

Figure 5. Comparison of measured 'Giant Resonance' with compouted cross-sections of atomic thorium using collective atomic models:LDRPA(Ref12) and RTDLDA(Ref15). The RTDLDA results indicate the presence of both ground and metastable state absorption.

is increased the f-waves begin to contract and the spectrum shows discrete structure overlaid on a giant resonance. Very suddenly all f-waves collapse into the inner well at which point the spectrum becomes a simple Rydberg series running up to the 5d ionization limits. We carried out a series of calculations on 5d - nl excitations with the Cowan suite of codes[16)]. We concentrated on more highly ionised thorium ions (4+ -> 6+) which have relatively simple ground states. The results are shown in Figure 6, where we show a comparison between a measured relative and computed absolute cross-sections.

It is clear that the spectrum is an admixture of all three ion stages and dominated by the main 5d - 5f transition which absorbs most of the 5d-spectrum's oscillator strength.

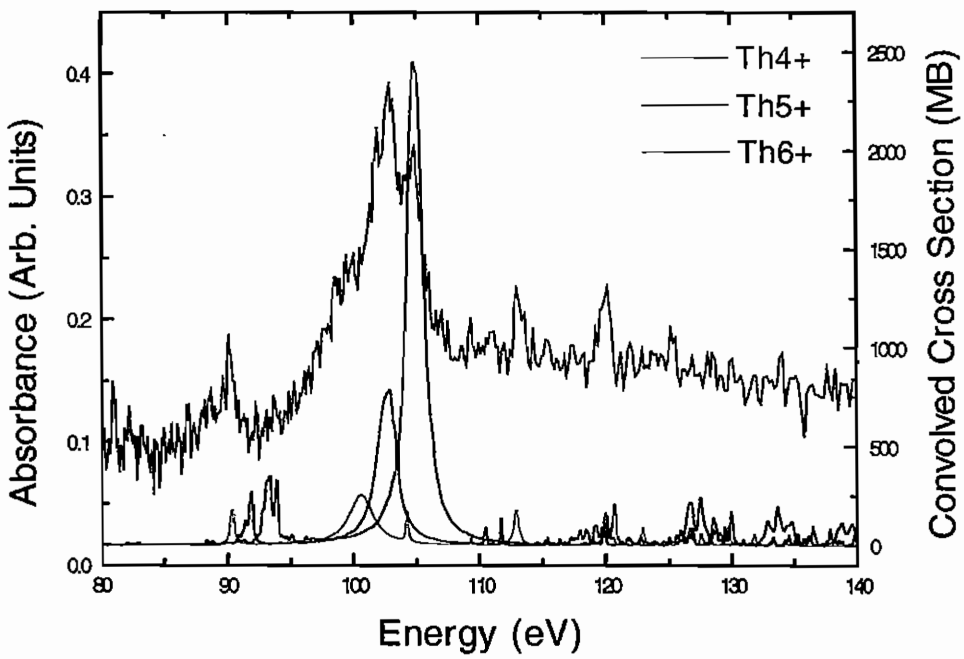

Figure 6. Photoabsorption spectrum at an interplasma delay of 20ns. The spectrum is compared with computed spectra for 4,5 & 6 time ionized thorium. The absorption is clearly due to predominantly five times ionized Th ions.

REFERENCES

1. A A Seraphin et al J.Appl.Phys **80**, 6429 (1996)
2. V Schmidt, Rep.Prog.Phys. **55**, 1483 (1992)
3. P K Carroll and E T Kennedy, Phys.Rev.Lett **38**, 1068 (1977)
4. J T Costello et al., Phys.Scr **T34**, 77 1991

5. E T Kennedy, J T Costello and J-P Mosnier, J.Electron.Spec.Relat.Phenom **79**, 283 (1996)

6. P K Carroll et al., IEEE,J.Q.Electron **QE-19**, 1807 (1983)

7. O Meighan et al., Appl.Phys.Lett **70**, 1497 (1997)

8. "Giant Resonances in Atoms, Molecules and Solids", Eds. J-P Connerade et al., (Plenum: 1987)

9. T B Lucatorto et al., Phys.Rev.Lett **47**, 1124 (1981)

10. U Koble et al., Phys.Rev.Lett **74**, 2188 (1995)

11. P K Carroll and J T Costello, J.Phys.B **20**, L201 (1987)

12. J C Kieffer and M Chaker, J.X-ray.Sci.Tech **4**, 312 (1994)

13. G Wendin, Phys.Rev.Lett **53**, 724 (1984)

14. P K Carroll and J T Costello, Phys.Rev.Lett **57**, 1581 (1986)

15. D A Liberman and A Zangwill, Comput.Phys.Commun **32**, 75 (1984)

16. R D Cowan, ftp://t4.lanl.gov/pub/cowan