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To cite this article: Asawari P Marathe et al 2007 J. Phys.: Conf. Ser. 80 012032

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New even parity autoionisation levels of U I

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Abstract. Even parity autoionisation levels of atomic uranium are investigated in the energy
region 51425 – 51760 cm \(^{-1}\) by two-colour three-photon ionisation spectroscopy in an atomic
beam using a time of flight mass spectrometer and in a hollow cathode discharge source using
photoionisation optogalvanic spectroscopy. Analysis of the photoionisation spectrum has
resulted in identification of 26 new levels and confirmation of 24 levels reported earlier. In
addition 9 odd parity levels in the energy region 34525 - 34860 cm \(^{-1}\) are confirmed.

1. Introduction

The study of autoionisation (AI) levels of heavy atomic systems is of considerable current interest
owing to their importance in basic atomic physics and in applications such as ultra-trace elemental
analysis [1] and isotope selective photoionisation [2]. Atomic uranium (U I) is a prototype of heavy
atomic systems, where the atomic structure is complicated by interaction of a large number of
optically active electrons. There have been a number of investigations on AI levels of U I using
various laser spectroscopic techniques, which have resulted in identifying a large number of even and
odd parity AI levels [3-10]. Despite this it is generally believed [10] that the present survey of AI
levels of U I is far from complete, particularly in the energy region that lies 1500 cm \(^{-1}\) above the first
continuum (5f\(^3\)7s\(^2\) \(^4\)I\(_{9/2}\), 49958.4 cm \(^{-1}\)). In this paper, we report our work on observation of new AI
resonances of uranium in 51530 – 51760 cm \(^{-1}\) region by two-colour photoionisation spectroscopy in an
atomic beam using a time of flight mass spectrometer (TOFMS) together with photoionisation
optogalvanic spectroscopy (PIOGS) in a hollow cathode discharge lamp (HCDL).

2. Experimental

A schematic representation of the experimental set-up is given in figure 1. It primarily consists of two
tunable pulsed dye lasers DL1 (of wavelength \(\lambda_1\)) and DL2 (of wavelength \(\lambda_2\)) pumped by a frequency
doubled high power Nd:Yag laser (~ 1 J/pulse @ 1064 nm), a high vacuum atomic beam generator
that is coupled to a linear TOFMS and a PIOGS set-up incorporating a uranium HCDL. The typical
line width and pulse duration of the dye lasers are ~ 4 GHz and ~ 10 ns respectively. The DL2 is
delayed by ~15 ns using an optical delay line. The atomic beam generator consists of a resistively
heated tantalum crucible maintained at ~ 2000 K and containing a few mg of natural uranium, and a
collimating structure to collimate the vapour to a desired degree. The typical number density of
uranium atoms in the laser-atom interaction region formed by the overlap of DL1 and DL2 with the
collimated atomic beam is ~10\(^7\) cm\(^{-3}\). The photoions generated in the interaction region are extracted
and accelerated in the TOFMS using dc electric fields of strengths 385 Vcm\(^{-1}\) and 500 Vcm\(^{-1}\).
respectively. After travelling a field free flight region of 70 cm, the photoions are detected by a channelltron detector. The output of the detector is amplified (gain ~ 100) and provided to a box car averager. For generating the two-colour photoionisation spectrum, $\lambda_1$ is held fixed and $\lambda_2$ is scanned in the desired wavelength range. In a similar manner a part each of DL1 and DL2 is used in a uranium HCDL and overlapped with the negative glow region of a uranium HDCL to obtain PIOGS signal which is recorded simultaneously with the TOFMS signal. It is found that the resonances in the photoionisation spectra obtained by these two independent techniques agree within $\pm 0.1$ cm$^{-1}$. Figure 1 also includes the wavelength calibration set-up for $\lambda_2$. The normal optogalvanic signal from a uranium HCDL is used for coarse calibration and interference fringes from a Fabry Perot etalon of free spectral range of 0.992 cm$^{-1}$ are used for fine calibration. While in general very rich two-colour photoionisation spectra can be generated by fixing $\lambda_1$ on a known resonance transition and scanning $\lambda_2$ over a wide wavelength range, this paper discusses a specific interesting case that corresponds to $\lambda_1$ tuned to the $5f^66d^7s^2 \, ^3L_6 \rightarrow 5f^66d^7s^7p \, ^3M_7$ transition at 591.538 nm (16900.38 cm$^{-1}$) and $\lambda_2$ scanned in the range 555–570 nm (17625 – 17960 cm$^{-1}$). The photoionisation spectrum therefore results from the two-colour three photon processes.

**Figure 1.** Schematic experimental set-up for two-colour photoionisation spectroscopy. DL1, DL2: tunable dye lasers, HCDL: hollow cathode discharge lamp, FP: Fabry Perot etalon, CH: high vacuum chamber for generating atomic beam, TOFMS: time of flight mass spectrometer, D: channeltron detector, AMP: pre-amplifier, BCA: box car averager, PD: photo diode, BD: beam dump, M: mirror, BS: beam splitter, ODL: optical delay line, PC: personal computer

### 3. Results and discussion

When $\lambda_1$ is fixed at 591.538 nm it gives rise to excitation of the $5f^66d^7s^7p \, ^3M_7$ level (16900.38 cm$^{-1}$) together with a two-photon resonant odd parity level at 33801.05 cm$^{-1}$ [7,10] owing to the band width of the laser (≈ 4 GHz). This results in a very rich photoionisation spectrum (spectrum-A) where the dominant contributions arise from two distinct two-colour three photon ionisation routes – (a) $\lambda_1 + 2\lambda_2$ wherein the odd parity levels in the energy region 34525 - 34860 cm$^{-1}$ reached by the combination $\lambda_1 + \lambda_2$ are photoionised by $\lambda_2$ and (b) $2\lambda_1 + \lambda_2$ wherein the two-photon resonant ($2\lambda_1$) level of odd parity at 33801.05 cm$^{-1}$ is excited to an AI level by $\lambda_2$.

In order to distinguish the resonances in the observed spectrum-A, the dependence of the strengths of the resonances on the detuning of $\lambda_1$ is studied. To this end an independent photoionisation
spectrum (spectrum-B) is obtained by fixing \( \lambda_1 = 591.338 \text{ nm} \) (16900.52 cm\(^{-1}\)) to bring the odd parity level at 33801.05 cm\(^{-1}\) to exact two-photon resonance and then scanning \( \lambda_2 \) as before. Photoionisation resonances corresponding to the routes (a) and (b) are respectively suppressed and enhanced in the spectrum-B in comparison to those in the spectrum-A. Since every resonance corresponding to the route (b) is associated with an AI level, these experiments provide an unambiguous identification of AI levels, which are listed in table 1.

**Table 1.** Even parity AI levels of uranium in the region 51425 –51760 cm\(^{-1}\) reached from 33801.05 cm\(^{-1}\) level.

| Present Work (cm\(^{-1}\)) | Literature (cm\(^{-1}\)) | Relative strength |
|-----------------------------|--------------------------|-------------------|
| 51425.4                     | 51425.7\(^a\)            | 28                |
| 51431.0                     | 51430.6\(^a\), 51431.2\(^b\) | 100               |
| 51457.0                     | 51456.9\(^a\)            | 66                |
| 51457.5                     |                          | 79                |
| 51458.4                     | 51458.3\(^a\)            | 56                |
| 51459.2                     | 51459.1\(^a\)            | 43                |
| 51459.6                     | 51459.6\(^a\)            | 29                |
| 51477.1                     | 51477.1\(^a\)            | 46                |
| 51478.5                     | 51478.5\(^a\)            | 33                |
| 51480.4                     | 51481.0\(^a\)            | 12                |
| 51483.4                     | 51483.9\(^a\)            | 15                |
| 51499.1                     | 51499.7\(^a\)            | 23                |
| 51500.9                     | 51500.2\(^a\)            | 15                |
| 51506.1                     | 51506.6\(^a\)            | 15                |
| 51510.8                     | 51510.8\(^a\)            | 65                |
| 51515.4                     | 51515.3\(^a\)            | 7                 |
| 51520.0                     | 51519.9\(^a\)            | 21                |
| 51523.7                     | 51524.0\(^a\)            | 15                |
| 51529.7                     | 51530.0\(^a\)            | 23                |
| 51532.4                     |                          | 8                 |
| 51534.0                     | 51533.9\(^b\)            | 47                |
| 51542.0                     | 51542.1\(^b\)            | 42                |
| 51549.4                     | 51549.5\(^a\)            | 20                |
| 51555.6                     | 51555.5\(^a\), 51555.6\(^b\) | 21              |
| 51581.2                     |                          | 20                |

| Present Work (cm\(^{-1}\)) | Literature (cm\(^{-1}\)) | Relative strength |
|-----------------------------|--------------------------|-------------------|
| 51583.6                     |                          | 10                |
| 51590.1                     |                          | 7                 |
| 51592.4                     |                          | 4                 |
| 51594.4                     |                          | 7                 |
| 51622.0                     |                          | 21                |
| 51629.4                     | 51629.7\(^b\)            | 15                |
| 51635.0                     |                          | 17                |
| 51659.6                     | 51659.7\(^b\)            | 19                |
| 51675.2                     |                          | 7                 |
| 51677.4                     |                          | 5                 |
| 51679.9                     |                          | 3                 |
| 51686.6                     |                          | 10                |
| 51690.1                     |                          | 6                 |
| 51691.1                     |                          | 5                 |
| 51703.2                     |                          | 5                 |
| 51714.2                     |                          | 20                |
| 51717.0                     |                          | 26                |
| 51725.1                     |                          | 33                |
| 51729.8                     |                          | 26                |
| 51741.3                     |                          | 36                |
| 51745.5                     |                          | 54                |
| 51748.6                     |                          | 74                |
| 51759.2                     |                          | 71                |
| 51759.7                     |                          | 56                |

\(^a\): Ref [7], \(^b\): Ref [10]
Of the 50 AI resonances observed in this work, 26 are being reported for the first time whereas 24 are found to be in good agreement with those reported in the literature. The strengths of these resonances, relative to the 33801.05 cm\(^{-1}\) \(\rightarrow\) 51431.0 cm\(^{-1}\) transition are also given in table 1. A comparison of the photoionisation spectra A and B also helps in the identification of 9 odd parity levels in the region 34525 - 34860 cm\(^{-1}\). These levels are presented in table 2 and they are in good agreement with earlier works.

### Table 2. Energy levels of uranium in the region 34525 –34860 cm\(^{-1}\) observed in the present work

| Present Work (cm\(^{-1}\)) | Literature (cm\(^{-1}\)) |
|-----------------------------|--------------------------|
| 34659.2                    | 34659.2\(^a\), 34659.2\(^b\), 34659.2\(^c\) |
| 34670.2                    | 34669.7\(^b\)             |
| 34675.8                    | 34675.4\(^b\), 34675.9\(^c\) |
| 34703.5                    | 34703.3\(^b\)             |
| 34718.2                    | 34718.1\(^a\), 34717.7\(^b\), 34718.0\(^c\) |
| 34746.4                    | 34746.3\(^a\), 34746.2\(^b\), 34746.1\(^c\) |
| 34765.1                    | 34764.8\(^a\), 34765.1\(^b\), 34765.0\(^c\) |
| 34798.0                    | 34798.0\(^a\), 34797.8\(^b\), 34797.7\(^c\) |
| 34803.6                    | 34803.7\(^a\), 34803.6\(^b\), 34803.7\(^c\) |

\(^a\): Ref. [3], \(^b\): Ref. [10], \(^c\): Ref. [11]

### 4. Conclusion

Autoionisation resonances of uranium are studied in the energy region 51425 – 51760 cm\(^{-1}\) by two-colour photoionisation spectroscopy in an atomic beam and a HCDL. These studies reveal 26 new autoionisation levels together with confirmation of 24 levels reported earlier. It may be noted here that the energy region investigated here lies \~ 1500 cm\(^{-1}\) above the first continuum and much of this region is still unexplored. Further work includes extending the energy region beyond 51760 cm\(^{-1}\) and also assigning the angular momenta (J) of the autoionisation levels unambiguously. Details of this work will be reported elsewhere.

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