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Determination of radiative lifetimes in neutral arsenic using time-resolved laser spectroscopy in the VUV region

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Abstract. Inspired by the recent Hubble Space Telescope first observations of arsenic in an astrophysical object we have determined the natural radiative lifetime of the upper states of two of the three resonance lines of As I. Short pulse laser excitation at 193.7 and 197.2 nm was achieved using different non-linear optical mixing schemes. Using time-resolved spectroscopy we obtain $\tau(4p^2 5s \ ^4P_{3/2}) = 4.3(5)$ ns and $\tau(4p^2 5s \ ^4P_{1/2}) = 4.5(5)$ ns yielding the absorption oscillator strengths $f_{\text{abs}}(193.7 \text{ nm}) = 0.123(17)$ and $f_{\text{abs}}(197.2 \text{ nm}) = 0.059(8)$.

Key words: arsenic – lifetime

Laser spectroscopy on elements such as arsenic has been hampered by the need for VUV laser pulses of short duration, which only now can be produced using non-linear optical mixing schemes. A further complication is that arsenic evaporates predominantly as a molecule, requiring means of dissociation to obtain free atoms. In the present work, we were able to handle these experimental difficulties. We have measured lifetimes for two resonance levels of As I, $4p^2 5s \ ^4P_{3/2}$ and $^4P_{1/2}$, and evaluated absorption oscillator strengths for the resonance lines at 193.7 and 197.2 nm. Below we describe our set-up and measurements, and discuss the results.

1. Introduction

Accidentally, arsenic was recently detected for the first time in an astrophysical object in the first scientific observation with the high-resolution Echelle spectrometer (GHRS) onboard the Hubble Space Telescope (Leckrone et al. 1991). The 193.7 nm resonance line connecting the $4p^2 5s \ ^4P_{3/2}$ level to the ground state $4p^3 \ ^4S_{3/2}$ was identified in the spectrum of Chi Lupi, which is a chemically peculiar B star of the “HgMn” type. This finding was made in a study of a 10 Å wide spectral region around the Hg II resonance line at 194.2 nm. The observation of arsenic spurred us to determine the lifetime of the $4p^2 5s \ ^4P_{3/2, 1/2}$ levels for providing the oscillator strength needed for an accurate abundance determination.

The first measurements of oscillator strengths in As I were reported in the monograph by Corliss & Bozman (1962). They measured oscillator strengths by studying the emission from an electric arc. The measured resonance lines of As I were near the sensitivity limit of the experimental set-up used. A more accurate method to obtain absolute oscillator strengths is to combine lifetime and branching ratio measurements. However, no cascade-free measurements of the radiative lifetimes for these states have been performed before. Lotrian et al. (1980) measured branching ratios in As I, and to obtain absolute oscillator strengths they used beam-foil measurements by Anderson et al. (1974). Semi-empirical calculations have been performed by Lawrence (1967) and Holmgren (1975).

2. Experimental set-up

The experimental set-up used in the present work is shown in Fig. 1. Arsenic was contained in a quartz cell that was baked out at 800 °C under high vacuum for 10 h prior to distillation of a small amount of arsenic into the cell. Arsenic vapour consists mainly of molecules but by thermal dissociation a minor fraction of free atoms can be produced. To promote this process the cell was differentially heated. One part of the cell (the stem) was kept at a lower temperature which determined the molecular vapour pressure, while the main part was kept at a higher temperature to promote thermal dissociation.

The laser radiation needed for the excitation was generated with slightly different set-ups. The 197 nm radiation, required to reach the $4p^2 5s \ ^4P_{1/2}$ state from the $4p^3 \ ^4S_{3/2}$ ground state, was produced by mixing of laser radiation at 281 and 622 nm in a BBO crystal. The energies of the pulses were approximately 10 mJ at 281 nm and 0.5 mJ at 622 nm. The 281 nm radiation was generated by frequency doubling of radiation from a Quantel Datachrome 5000 dye laser, operated on Rhodamine 6G dye which was pumped by the second harmonic at 532 nm from a Quantel YG:580 Nd:YAG laser. The radiation at 622 nm was generated by a Quanta-Ray PDL dye laser with DCM dye, where the oscillator was pumped by radiation at 337 nm from a VSL 337ND nitrogen laser from Laser Science. The pulses from the nitrogen laser were 3 ns long which induced pulses of equal length in the oscillator. The amplification steps in the Quanta-Ray system were pumped by the second harmonic radiation from the Nd:YAG laser with a pulse length of 10 ns, but the length of the resulting pulse was limited by the duration of the pulse from the oscillator. Likewise, in the final mixing process the short pulse at 622 nm determined the length of the resulting pulse at 197 nm to approximately 3 ns.

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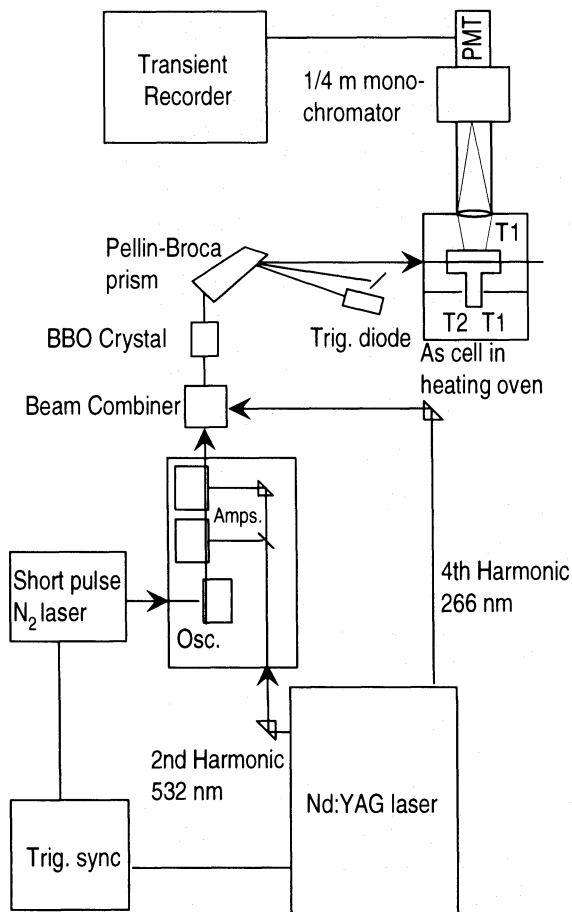


Fig. 1. Experimental set-up used in time-resolved measurements on arsenic atoms

Supplementary data were recorded with radiation at 197 nm generated by quadrupling of the frequency of the fundamental beam at 788 nm from the Quanta-Ray dye laser. This was accomplished by first doubling the frequency in a KDP crystal and subsequently mixing the fundamental and the second harmonic beam in a BBO crystal resulting in a beam at 263 nm. Finally, this beam was again mixed with the fundamental beam in a further BBO crystal to obtain the desired radiation at 197 nm. This set-up had the disadvantage of a longer pulse determined by the pulse length of the pumping Nd:YAG laser, which was about 10 ns. Fortunately, the Nd:YAG laser pulse had a three-peak temporal structure with a dominating central peak. With the oscillator of the dye laser operating near threshold only the central peak caused lasing, thereby shortening the resulting pulse. In addition the pulse was shortened by the three non-linear processes in the crystals, which enhanced the peak of the pulse with regard to the weaker parts at the ends. The resulting pulse at 197 nm had a pulse length of approximately 4 ns.

The radiation at 194 nm required to reach the $4p^2 5s \ ^4P_{3/2}$ state was generated by mixing the fourth harmonic of the Nd:YAG laser at 266 nm with radiation at 713 nm from the Quanta-Ray dye laser in a BBO crystal. The energy of the 266 nm pulses was about 20 mJ while the pulse energy at 713 nm was less than 0.5 mJ. The Quanta-Ray dye laser was run on LDS 698 dye dissolved in DMSO and pumped in the same way as in the

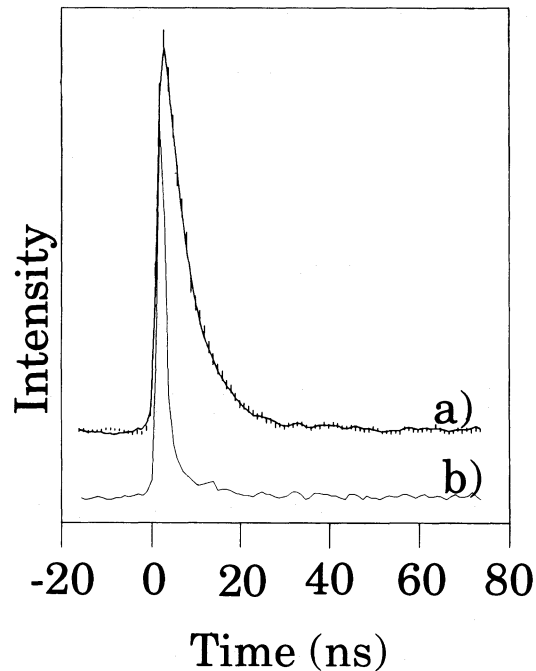


Fig. 2. Recording of (a) fluorescence decay from the $4p^2 5s \ ^4P_{3/2}$ state of neutral arsenic and (b) laser pulse as seen by the detection system

197 nm set-up to obtain short pulses. Also, for this state complementary data were recorded with frequency quadrupled radiation from the Quanta-Ray dye laser in conformity with the second scheme for the $4p^2 5s \ ^4P_{1/2}$ state.

The detection of the fluorescence decay was made using a Hamamatsu R1564U micro-channel photomultiplier tube connected to a Tektronix DSA 602 Transient Digitizer. The decay from the $4p^2 5s \ ^4P_{3/2}$ state was monitored by observing the fluorescence on the 245 nm line to the $4p^3 \ ^2D_{3/2}$ level, while the $4p^2 5s \ ^4P_{1/2}$ state was monitored on the 249 nm line to the same level. Lifetime evaluation was performed on an IBM-compatible personal computer by fitting the experimental decay curve to a convolution of an exponential decay and the recorded laser pulse. Such a procedure is valid in a single-step excitation process where the atomic detector response is linear. These conditions are fulfilled in our experiments. Fig. 2 shows a typical fluorescence curve for the $4p^2 5s \ ^4P_{3/2}$ state and a recording of the shape of the laser pulse as seen by the detection system. A fit of a convoluted

Table 1. Radiative lifetimes of As I $4p^2 5s$ states

State	Lifetime value (ns)					
	Experiment			Theory		
	This work	[1]	[2]	[3]	[4] DV	[4] DL
$^4P_{1/2}$	4.5(5)	3.6(4)	2.2	8.4	8.1	4.7
$^4P_{3/2}$	4.3(5)	3.8(4)	1.3	8.2	8.1	4.8

References: [1] Anderson et al. (1974); [2] Corliss et al. (1962); [3] Lawrence (1967); [4] Holmgren (1975).

Table 2. Absorption oscillator strengths of As I resonance lines

Line (nm)	Absorption oscillator strength							
	Experiment					Theory		
	This work	[1, 2]	[3]	[4]	[5]	[6]	[7] DV	[7] DL
197.2	0.059(8)	0.074(10)	0.07	—	—	0.03	0.03	0.06
193.7	0.123(17)	0.14(2)	0.05	—	—	0.06	0.06	0.11
f_1/f_2	2.1(5)	1.9(4)	0.71	1.7	2.5	2.0	2.0	1.8

References: [1] Lotrian et al. (1980); [2] Andersen et al. (1974); [3] Corliss et al. (1962); [4] L'vov (1972); [5] Muradov (1977); [6] Lawrence (1977); [7] Holmgren (1975).

curve to the experimental decay curve is included, exhibiting very small residues.

3. Measurements and results

Signals were recorded at temperatures ranging from 260 to 320 °C at the coldest point of the cell, while the temperature at the hot part varied between 350 and 500 °C. No dependence between lifetimes and temperatures was observed in the temperature range studied. Thus, we deduce that neither multiple scattering nor collisions with molecules influence our experiments.

The lifetimes were measured to be 4.3(5) ns for the $4p^25s^4P_{3/2}$ state and 4.5(5) ns for the $4p^25s^4P_{1/2}$ state. The quoted error bars comprise statistical scattering as well as an estimated uncertainty in the deconvolution procedure. In Table 1 these values are compared with measured beam-foil values (Andersen et al. 1974), values derived from oscillator-strength measurements (Corliss & Bozman 1962) and results from calculations (Lawrence 1967; Holmgren 1975).

We note that our measured lifetime values are slightly higher than the beam-foil data (Andersen et al. 1974) but lower than the values calculated (Holmgren 1975) by the optimized Hartree–Fock–Slater method using the dipole-velocity (DV) or the dipole-length (DL) operator. We conclude that there is agreement within the experimental errors between two experimental and one theoretical set of data. Using the radiative lifetime values and branching ratios, derived from (Lotrian et al. 1980), we obtain the oscillator strengths for the As I resonance lines, that are given in Table 2.

From Table 2 we can again conclude that the best agreement for absolute oscillator strengths is obtained between our results, the Lotrian et al. data and Holmgren's theoretical calculations

using the DL operator. For the relative oscillator strengths there are two experimental investigations. Absorption measurements have been performed by L'vov (1972) and by Muradov (1977). We note that our ratio of the two f -values, f_1/f_2 , as well as that by Lotrian et al. fall between the ratios obtained in the quoted absorption measurements. We also get a better agreement with the calculations.

For the determination of arsenic abundances we recommend the oscillator strengths obtained from this work based on our new lifetime measurements and the branching ratios given by Lotrian et al. (1980).

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