

Determination of radiative lifetimes of neutral sulfur by time-resolved vacuum-ultraviolet laser spectroscopy

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Time-resolved laser spectroscopy was used to measure radiative lifetimes of eight excited states of neutral sulfur in a laser-produced plasma. Excitation from the atomic ground state was performed with pulsed vacuum-ultraviolet (126–140 nm) radiation generated by resonant sum-difference four-wave mixing in krypton gas. The lifetimes of the $3p^3ns\ ^3S^o$ ($n=5-7$) and $3p^3nd\ ^3D^o$ ($n=4,5$) states were measured and are presented together with data available from literature. Relativistic Hartree-Fock calculations of radiative lifetimes were also performed for states where experimental data were available. [S1050-2947(97)04403-X]

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I. INTRODUCTION

Sulfur is a biological trace element and its presence in the human body is important for life. On the other hand, some sulfur compounds are toxic. Since they are frequently products of human activities, the amount of sulfur compounds in the environment is increasing with time. Since a large number (~ 80) of sulfur atomic lines has been observed in the spectra of the sun as well in other astrophysical objects and in view of the high cosmic abundance of sulfur, astrophysicists are interested in refining the abundance values of this element. However, only a small number of the oscillator strengths needed are available from literature. In the latest theoretical calculation [1] of sulfur oscillator strengths, investigations performed by different authors are reviewed. Most papers concern theoretical calculations, and accurate experimental data are needed for evaluation of their accuracy. On the other hand, the real situation is opposite; for example, in the evaluation of the sulfur abundance in ζ -Ophiuchi [2] it was suggested to use a multiplicative correction coefficient of about 1.2–1.6 for the experimental data.

Part of the S I energy level diagram is shown in Fig. 1. To our knowledge, no attempt has been made up to now to predict theoretically the radiative lifetimes of the high excitation ($n \geq 5$) levels experimentally investigated in the present work. The complexity of the S I spectrum and the huge number of interacting configurations have apparently prevented such attempts. The results previously published were obtained in the framework of monoconfigurational approximations [3–7] or were intermediate-coupling results concentrating on transitions originating from the ground state [8–11]. The most extensive set of results are due to Aymar [12], Kurucz and Peytremann [13], and Fawcett [14] or were obtained in LS coupling in the framework of the opacity

project [15]. For these reasons, we have undertaken an alternative calculation of radiative lifetimes for the experimentally studied states in the present work.

Oscillator strengths have been measured in emission by using a wall-stabilized arc [16–19] and a shock tube [20]. Such measurements suffer from two major problems: (i) the evaluation of the concentration of atoms in excited states, and (ii) the calibration of the spectral sensitivity of the detection system, especially in the VUV spectral region. Possible influences of reabsorption and radiative trapping in the light source have to be considered. Only for few excited states have lifetime measurements been performed by using the phase-shift method following electron excitation [21,22], the high-frequency deflection technique [23], and time-of-flight spectroscopy [24]. No data from beam-foil experiments are available for neutral sulfur. First lifetime measurements by using two-photon UV laser excitation have just been performed [25]. All experimental lifetime data are given in Table I. Upper limits of the lifetime values evaluated from oscillator strength measurements are also listed. Where it was possible the results of the theoretical calculations are represented.

Lifetimes for a range of elements have been measured at

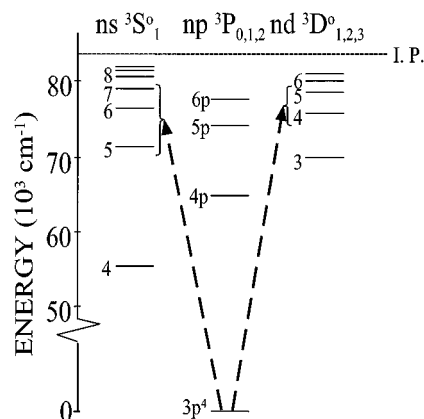


TABLE I. Experimentally determined radiative lifetimes from literature and data from the present work. An asterisk denotes unreliable results (cancellation effects).

Level	Energy (cm ⁻¹)	This work HFR	Lifetime (ns)	
			This work (expt.) and [25]	Expt. other
4s ⁵ S ₂ ^o	52624			9200, ^a 11 900, ^e >10 000, ^f 27 000 ^g
4s'' ³ P ₀ ^o	77136	2.9		
4s'' ³ P ₁ ^o	77150	2.9		2.8(0.3), ^b <1.2 ^e
4s'' ³ P ₂ ^o	77181	2.8		
4s' ¹ D ₂ ^o	69238	1.6		1.5(0.3), ^d <1.7 ^e
4s ³ S ₁ ^o	55331	1.4		1.5(0.3), ^d 1.4 ^e
5s ³ S ₁ ^o	71351	3.9	7.1(0.5)	<6.4 ^e
6s ³ S ₁ ^o	76721	8.4	17.7(1.1)	<5 ^e
7s ³ S ₁ ^o	79185	19.4	35.6(4.0)	
4p ³ P ₀	64891	27.3	46.1(1.0)	33(12) ^c
4p ³ P ₁	64889	27.3	46.1(1.0)	33(12) ^c
4p ³ P ₂	64893	27.2	46.1(1.0)	33(12) ^c
4p ³ P ₂	79376	42.9		53.5(4.0) ^a
5p ³ P ₂	74269	103.4	188(13)	185(15) ^a
5p ⁵ P ₃	73921	103.9		615(50), ^a <97(30) ^c
6p ⁵ P ₃	77856	207.9		265(20) ^a
7p ⁵ P ₃	79786	269.0		415(25) ^a
3d ³ D ₁ ^o	70165	3.4		
3d ³ D ₂ ^o	70166	3.4		2.1(0.3), ^d <1.5 ^c
3d ³ D ₃ ^o	70174	3.3		
4d ³ D ₁ ^o	75952	5.7	12.6(1.3)	11.4(1.5), ^b <8.5 ^e
4d ³ D ₂ ^o	75952	5.6	12.5(0.7)	11.4(1.5), ^b <8.5 ^e
4d ³ D ₃ ^o	75957	5.5	12.9(1.1)	11.4(1.5), ^b <8.5 ^e
5d ³ D ₁ ^o	78693	8.3	41(3)	
5d ³ D ₂ ^o	78691	8.2		
5d ³ D ₃ ^o	78692	8.0	40(4)	
6d ⁵ D ₄ ^o	79992	*		260(20) ^a
7d ⁵ D ₄ ^o	80995	473		505(40) ^a
3p ⁵ ¹ P ^o	81740			17(2), ^d <10.8 ^e

^aReference [23].

^bReference [22].

^cReference [17].

^dReference [21].

^eReference [18].

^fReference [37].

^gReference [38].

the Lund High-Power Laser Facility [26], by using VUV laser excitation. Different nonlinear techniques such as Raman shifting in hydrogen [27], higher-order harmonics generation in a rare-gas jet [28], and sum-difference four-wave mixing in krypton [29] have been applied for VUV light generation. For the generation of laser light in the 120–140 nm region, corresponding to sulfur resonance lines, the last of the above-mentioned methods is chosen.

In this paper we report on lifetime measurements for eight excited states of sulfur atoms measured from the recorded fluorescence decay after VUV laser excitation in a laser-produced plasma. This method has earlier been used for refractory elements and ionic states [30,31].

The number of investigated levels was limited by several factors. The upper limit of lifetime values obtainable by this method is defined by two factors, collisional deexcitations in

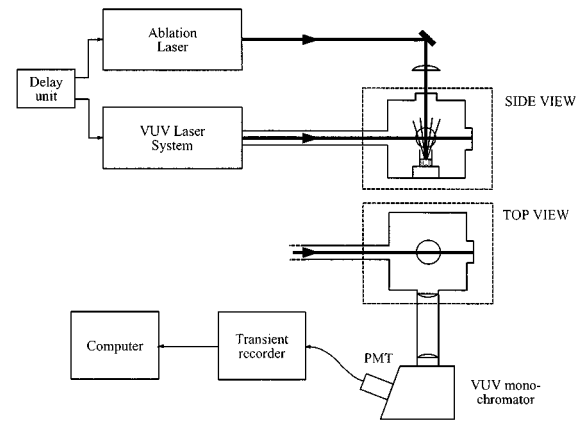


FIG. 2. Experimental setup for time-resolved laser spectroscopy.

the plasma and flight-out-of-view effects. As was demonstrated in [25], lifetimes below 200 ns can be measured without systematic shifts. The lower limiting factor for short lifetimes is the duration of the excitation laser pulse and as demonstrated in [27], a value down to 5 ns can be accurately measured. Many resonance levels in sulfur are more short-lived. Another limiting factor in sulfur is that quintet states are not available for laser excitation from the ground triplet. A solution for quintet *P* states could be laser excitation from the high-lying long-lived (9 μs) ⁵S₂^o state. On the other hand, the higher quintet states are expected to be long-lived, and influences of collisions and flight-out-of-view effects have to be investigated.

II. EXPERIMENTAL SETUP

The experimental setup used in the present experiments is shown in Fig. 2. To obtain the required VUV radiation, nonlinear frequency mixing in several steps was performed. Two dye lasers were pumped by two neodymium-doped yttrium aluminum garnet (Nd:YAG) lasers. One of them was adjusted to 637.65 nm giving 212.55 nm as the third harmonic, corresponding to the two-photon resonance in krypton. The third-harmonic radiation was produced by mixing of frequency-doubled light (in a KDP crystal) with the fundamental beam in a BBO crystal. A mechanically compressed crystalline quartz slab was used to obtain parallel polarization planes for the fundamental and frequency-doubled laser beams before the BBO crystal. To produce 126–140 nm wavelengths, the UV laser beam was merged with a visible 430–700 nm laser beam from a second dye laser. The merged beams were focused by an achromatic lens into a krypton-gas cell kept at 10–100 mbar. The output side of the cell was attached to a vacuum system and separated from it by a LiF lens. The VUV radiation was extracted from the outgoing beams by using a LiF monochromator and was made to cross a laser-produced plasma. Fluorescence light was collected by CaF₂ lenses perpendicularly to the laser and was focused to the slit of a vacuum monochromator (Acton model VM 502) and finally detected by a photomultiplier tube (Hamamatsu R1220). The data acquisition and evaluation were performed by using a digital oscilloscope

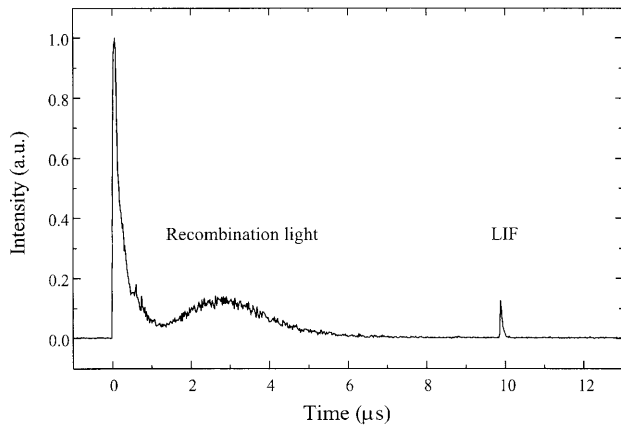


FIG. 3. Detected light intensity around 126 nm as a function of time after the ablation pulse. The signal at about 10 μs is laser-induced fluorescence from the $3p^3 7s^3 S_1^o$ state. Signals before 6 μs are recombination light from ions and atoms.

(Tektronic model DSA 602) and a personal computer. More details about this part of the experimental setup are given elsewhere [29].

A laser-produced plasma was used as a sulfur atom source. Pulses from a Nd:YAG laser beam were focused into lead sulfide powder. The powder was confined in a metallic container. A pulse energy of 10–30 mJ caused ablation of the powder, which was splashing at the laser pulse impact. To maintain a smooth target surface for the next laser pulse, the container was shaken by an electromagnet at a frequency in resonance with mechanical vibrations of the magnet (container) assembly. More details of the plasma source are given in [31].

All three Nd:YAG lasers were triggered from a common unit (Stanford Research System model 535). This allowed us to tune time delays between all three lasers and to obtain optimal time overlap of two of them for frequency mixing, and to change the time delay between the light pulses used for atomization and excitation.

III. MEASUREMENTS AND RESULTS

In order to obtain reliable radiative lifetime values we used time-resolved laser spectroscopy ensuring appropriate experiment conditions. A sufficient magnetic field has to be applied to wash out quantum beats from magnetic sublevel splittings due to the Earth's magnetic field. To make sure that the lifetimes are not affected by radiation trapping and collisions in the laser-produced plasma, we performed experiments with different time delays (7–20 μs) between the atomization laser pulse and the excitation laser. The intensity of light around 126 nm is shown as recorded as a function of time; see Fig. 3. In the beginning the emission from the plasma corresponding to sulfur ions with different ionization degrees are observed. A broad maximum at 3 μs is emission from neutral sulfur atoms, moving slower than ions. The peak at 10 μs is fluorescence light from the $3p^3 7s^3 S_1^o$ state following the laser excitation. In our measurements the shorter delay time was limited by plasma light emission after the first Nd:YAG laser pulse. The longest delay time was determined by decreasing the atomic concentration and the corresponding signal level after the laser excitation; see Fig.

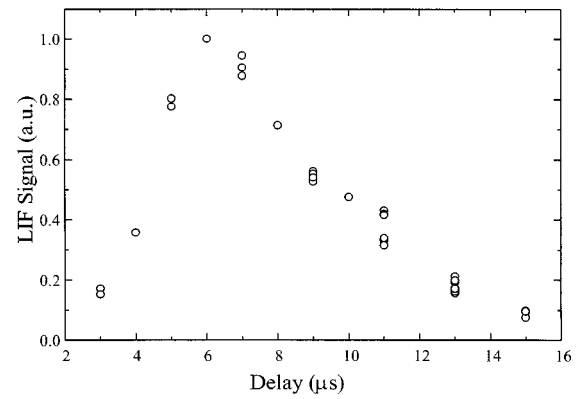


FIG. 4. Laser-induced fluorescence intensity from the $3p^3 7s^3 S_1^o$ state as a function of the delay time for the VUV laser with respect to the ablation pulse.

4. Here the maximum intensity of the fluorescence light after the laser excitation is plotted versus delay time. The intensity is related to the concentration of sulfur atoms in the ground state.

For short-lived states (<10 ns) the laser pulse was recorded to allow deconvolution calculations on the experimental curve in order to obtain the true exponential decay. One prerequisite for the deconvolution procedure is that the laser intensity is kept low enough to avoid transition saturation. For longer-lived states it was possible to evaluate the lifetime values directly from recorded decay curves, by fitting to an exponential, for times late enough (>10 ns) to be outside of the duration of laser pulse. Recordings of the laser pulse and fluorescence decay curves for the states of the $3p^3 ns^3 S_1^o$ ($n=5-7$) sequence are shown in Fig. 5. For each level about 30–40 decay curves for different delay times between the atomization and excitation laser pulses were recorded using signal averaging of many individual transients. A plot of evaluated lifetimes versus delay time for the $3p^3 5s^3 S_1^o$ state is given in Fig. 6, and it shows no depen-

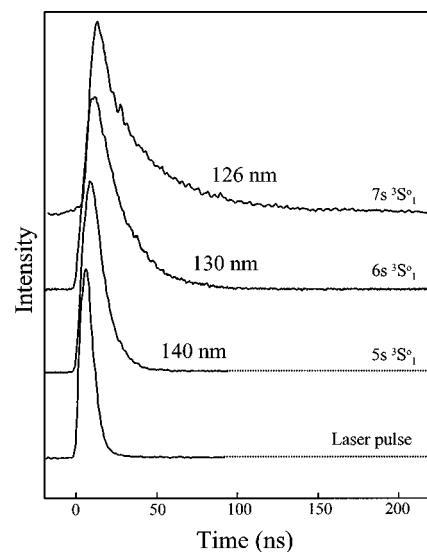


FIG. 5. Recordings of the fluorescence decay in the $ns^3 S_1^o$ ($n=5,6,7$) sequence of sulfur. The excitation pulse curve is also indicated.

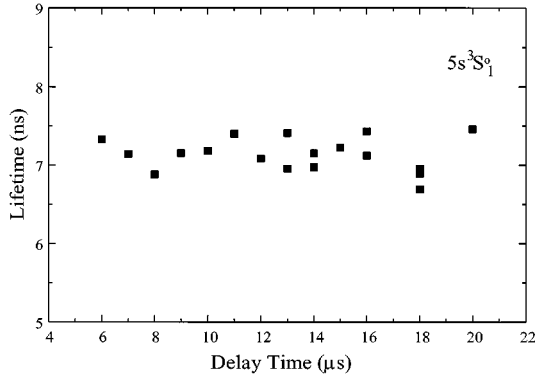


FIG. 6. Measured radiative lifetime of the $5s\ 3S_1^o$ state as a function of delay time.

dence on delay times for our experimental conditions. An arithmetical mean value is calculated and given in Table I as the radiative lifetime for each state. Two standard deviations are taken as error bars. Possible systematic shifts are not included in the error bars, but we tried to avoid such shifts by finding the appropriate experimental conditions. In the lifetime range between 5 and 50 ns, data have been obtained by using different atomization and detection techniques in our earlier works [25] and [27], and good agreement was observed.

As we see from Table I, comparisons of lifetimes can be performed only for a few levels. We note quite good agreement between our data and the phase-shift measurement [22] for the $4d\ 3D^o$ state, as well as between our data [25] and high-frequency-deflection data [23] for the $5p\ 3P_2$ state. We see that lifetimes evaluated from oscillator strengths [17,18] are shorter; corrections must be applied as was already pointed out in [2].

We have plotted the lifetimes in the $3p^3ns\ 3S_1^o$ ($n=4-7$) sequence versus the effective quantum number n^* on a log-log scale in Fig. 7. The straight line shows an n^{*3} dependence fitted to the $6s$ state. As we see, the lifetimes for $n=5-7$ follow a cubic law within error bars. For the lifetime of the $4s$ state [1.5(0.3) ns measured by using electron excitation [21]] the deviation is larger than the error bars. 2.2 ns would fit a cubic dependence. The lifetime for this state evaluated from oscillator strength measurements [18] is 1.4

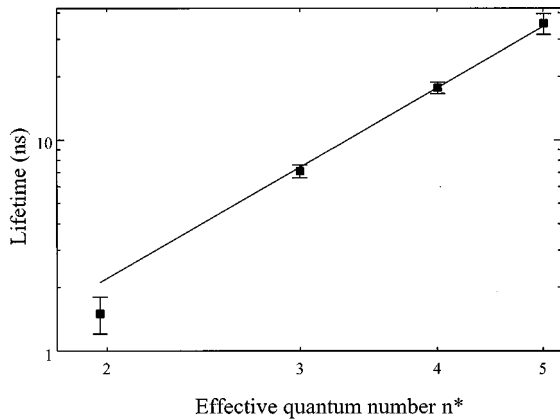


FIG. 7. Radiative lifetimes in the $ns\ 3S_1^o$ ($n=4-7$) sequence of sulfur as a function of effective quantum number on log-log scales.

ns. However, we observed systematically shorter values for other levels in that work [18].

IV. THEORETICAL RESULTS

The relativistic Hartree-Fock (HFR) code of Cowan [32] has been adopted for the calculations. It was combined with a least-squares fitting of the calculated eigenvalues of the Hamiltonian to the experimental levels in order to optimize the Slater parameters. The average energies (E_{av}) and the Slater integrals (F^k and G^k) were adjusted to reproduce the experimental levels compiled by Martin *et al.* [33]. The F^k , G^k , and R^k integrals, not optimized in the fitting procedure, were scaled down by a factor of 0.85 in agreement with a well known procedure [32]. For the sake of conciseness, the optimized parameters will not be reproduced here, but they are available upon request from the authors. In order to consider the most important relativistic and configuration interaction effects, the following configurations were explicitly introduced in the model:

$$\begin{aligned} &3s^23p^4 + 3s^23p^3np \quad (n=4-7) + 3s^23p^3nf \quad (n=4-7) \\ &+ 3p^6 + 3s3p^4nd \quad (n=3-5) + 3s3p^44s + 3s3p^45s \\ &+ 3p^54p + 3p^55p + 3p^54f + 3p^55f \end{aligned}$$

and

$$\begin{aligned} &3s^23p^3ns \quad (n=4-7) + 3s^23p^3nd \quad (n=3-7) + 3s3p^5 \\ &+ 3s3p^44p + 3s3p^45p + 3s3p^44f + 3s3p^45f \\ &+ 3p^5nd \quad (n=3-5) + 3p^54s + 3p^55s \end{aligned}$$

for the even and odd parities, respectively.

The theoretical lifetime values are reported in Table I, where they are compared with experiment. For the low-lying levels, particularly for $4s''\ 3P^o$, $4s'\ 1D^o$, and $4s\ 3S^o$, and to a lesser extent for $4p\ 3P$ and $4p'\ 3P$, the HFR results are in reasonable agreement with experiment. The discrepancies, however, do increase substantially when n , the principal quantum number, is increased. This gives an indication that some relevant configuration interaction effects have probably not been adequately considered in the present model. In particular, interactions with higher Rydberg states ($n>7$) and with the continuum should be further investigated. The scaling down of the Slater integrals, which is expected to catch the largest part of such effects, appears insufficient to correctly mimic the interactions with higher energy configurations. However, it should be emphasized that a detailed consideration of these correlation effects would require more powerful computer capabilities than the ones used in the present work.

In addition, as pointed out by Martin *et al.* [33], some problems arise concerning the assignments of some levels situated above the first ionization limit. More particularly, the assignments of the $(^2D^o)nd\ 3S^o$ and $(^2D^o)ms\ 3D^o$ levels appear to be ambiguous for $n+2=m\geq 5$, none of the calculations used by Martin *et al.* [33] in their compilation being sufficiently accurate to warrant definitive interpretation of the observed spectra. This imposes some limitations to the fitting procedure used in the present work.

TABLE II. Comparison between our experimental lifetimes (in ns) and literature theoretical data. CA denotes Coulomb approximation, FO denotes first order approximation, CI denotes configuration interaction, HF denotes Hartree-Fock method, DL denotes dipole length approximation, DV denotes dipole velocity approximation, ECI denotes extended configuration interaction, HFR denotes relativistic Hartree-Fock, SST denotes SUPERSTRUCTURE code.

Level	[8]	[9]	[12]		[11]			[1]	[35]	This work and [25]			
	IC	CA	FO	CI	HF, DL	HF, DV	ECI, DL	ECI, DV	HFR	SST	HFR	Expt.	
$4s\ ^3S^o$	1.4	3.4	1.4	1.5	2.7	1.9	2.3	2.2			1.4	1.4	2.2
$4s\ ^1D^o$	1.5	3.5	1.5	1.6	2.4	2.2	2.7	2.6				1.6	
$5s\ ^3S^o$			4.9		12	22	7.5	6.1	<40	<51	6.2	3.9	7.1
$4d\ ^3D^o$					13	10	11	14	<89	<87	10.7	5.6	12.7
$4p\ ^3P$			39	39					40	42	45	27	46.1
$5p\ ^3P$									280	170		103	188

As a consequence of these considerations, the present HFR results must still be considered as preliminary and further analysis will be carried out in the future in order to try and improve the present theoretical results.

V. DISCUSSION

For certain levels ($n \leq 5$) it is possible to compare our data with results of earlier theoretical calculations (Table II). We see that the theoretical data deviate from investigation to investigation. The large deviation between data obtained in [8] and [9] is due to the different methods used for the normalization to an absolute scale. In [8] the experimentally measured lifetimes from [17] are used: therefore, there is good agreement between those works. In [9] the quantum defect obtained from experimental energy levels [34] is used. The energy levels have been revised in [33] and slightly deviating values from the earlier data are obtained. In the evaluation of effective quantum numbers we use the energies from [33], which are indicated in Table I. The results obtained by the configuration interaction method [12] have better agreement with [8], but for the $5s\ ^3S^o_1$ level the value is smaller than our experimental value. The data obtained by the extended configuration-interaction method [11] are intermediate between the results from [8] and [9]. They are cal-

culated only for the transitions to the ground level, but if we combine them with weaker transitions to higher states calculated in [2], we obtain lifetimes for the $5s\ ^3S^o_1$ and $4d\ ^3D^o_1$ states in rather good agreement with our experimental values. For the $4p\ ^3P$ and $5p\ ^3P$ states the best agreement with our data is observed for calculations by using the SUPERSTRUCTURE computer code [1].

Future lifetime measurements in sulfur would benefit from using a short-pulse laser in the 120–160 nm region. We have observed strong fluorescence signals on a number of the resonance lines, but the lifetimes are too short to be evaluated using our excitation pulse. The metastable singlet states are also populated and strong fluorescence is observed by excitation at 167 nm corresponding to the $3p^4\ ^1D_2 - 3p^3 4s\ ^1D^o_2$ transition. Another interesting state for investigation is $4s\ ^5S^o_0$, which is very long-lived but with strongly conflicting experimental lifetime values as it was already mentioned in [36]; see Table I.

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