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LIFETIMES AND OSCILLATOR STRENGTHS FOR ULTRAVIOLET TRANSITIONS IN NEUTRAL SULFUR

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ABSTRACT

A consistent set of f -values for vacuum ultraviolet lines of S I is deduced from a combination of laser lifetime measurements and theoretical branching ratios. The new results are compared with results deduced from absorption by interstellar gas toward ζ Ophiuchi, and good agreement is observed for most transitions.

Subject headings: atomic data — ISM: abundances

1. INTRODUCTION

Absorption from the ground state of neutral sulfur arises from a large number of lines at far-ultraviolet wavelengths (see Morton 1991). The quality of data from the *Hubble Space Telescope* (*HST*) allows detection of many of the weakest transitions from diffuse interstellar clouds (e.g., Federman & Cardelli 1995; Zsargó, Federman, & Cardelli 1997). Federman & Cardelli (1995) analyzed S I absorption from interstellar gas toward ζ Ophiuchi in order to obtain a set of self-consistent f -values. Their analysis was based on precisely known experimental f -values for the multiplet at 147.9 nm (Doering 1990; Beideck et al. 1994), from which the column density and Doppler parameter (b -value) were derived. The f -values for the other lines were adjusted until each line yielded the same column density and b -value. For the stronger lines, the accuracy of the inferred f -values is limited by the precision of the experimental results, namely, 10%. The f -values for the weaker lines are accurate to about 20%.

Since the completion of the analysis by Federman & Cardelli (1995), additional experimental data on S I became available (Berzinsh et al. 1997). S I has also been the subject of recent theoretical investigations (Chen & Msezane 1997; Tayal 1997). The main purpose of this paper is to provide a comparison of the lifetimes inferred from the f -values of Federman & Cardelli (1995) and those of Berzinsh et al. (1997), supplemented by new lifetime values reported here for the first time. These empirical results are also compared with recent theoretical determinations based on large-scale computations. New relativistic Hartree-Fock (HFR) calculations, which were performed to elucidate the causes for differences (when they occur) between the laboratory and astronomical results on the one hand and the theoretical ones on the other, are also described.

2. NEW EXPERIMENTAL RESULTS

The lifetimes of the $6d^3D^o$ and $8s^3S^o$ levels have been measured in the present work by time-resolved VUV laser spectroscopy. The two levels were populated by a three-photon excitation process (see the level diagram of Fig. 1),

and the radiative lifetimes were deduced from the observation of the light intensity decay of the lines originating from these levels. The details of the experiment will be described elsewhere (Li et al. 1998), and we just sketch out the basic ideas of the method used.

The target was formed by a laser-produced plasma containing sulfur atoms. The atoms were created by the dissociation of lead sulfide molecules irradiated by a 10–30 mJ frequency-doubled Nd:YAG pulsed laser. The laser light was focused on lead sulfide powder contained in a little cup holder which was mechanically shaken to guarantee a smooth target surface for each pulse. In a first step, a two-photon excitation of the $4p^3P$ level from the ground state was used (see Fig. 1). To obtain the required UV radiation at ≈ 308 nm, the frequency-doubled output of a Nd:YAG laser was used to pump a DCM dye laser tuned at about 616 nm. That light was subsequently frequency doubled in a KDP crystal to produce a 10 Hz, ≈ 7 ns pulse of 25 mJ power. The laser light was tuned in resonance with the $3p-4p$ transition by adjusting the dye-laser wavelength. The population of the $4p$ levels was monitored by the induced fluorescence on the $4s-3p$ line at 180.7 nm.

In a second step, the levels $6d^3D^o$ and $8s^3S^o$ were excited from the levels $4p^3P$. The radiation at about 650 nm was obtained from a dye laser pumped by a frequency doubled Nd:YAG laser. The tuning in resonance was monitored by looking at the intensity of the signal produced by the decay of the upper level in the VUV (≈ 125 nm) region. The target holder was placed at the center of a magnet in order to eliminate possible quantum beats. The two excitation laser beams were horizontal and perpendicular to the magnetic field. The fluorescence was observed along the magnetic field axis by using two MgF_2 lenses focusing the source light on the entrance of a 20 cm normal-incidence vacuum spectrometer. The light was detected by a solar blind photomultiplier, the signal of which is recorded, in synchronization with the pulse arrivals, by a fast transient recorder (1 ns channel $^{-1}$ timescale, 1024 channels). The spectrometer acts as a wavelength filter (1 mm slits, 4 nm resolution) and allows discrimination between different signals (i.e., between the 180 nm and the 125 nm lines). Each decay curve was recorded by averaging 4096 laser shots, and the data were

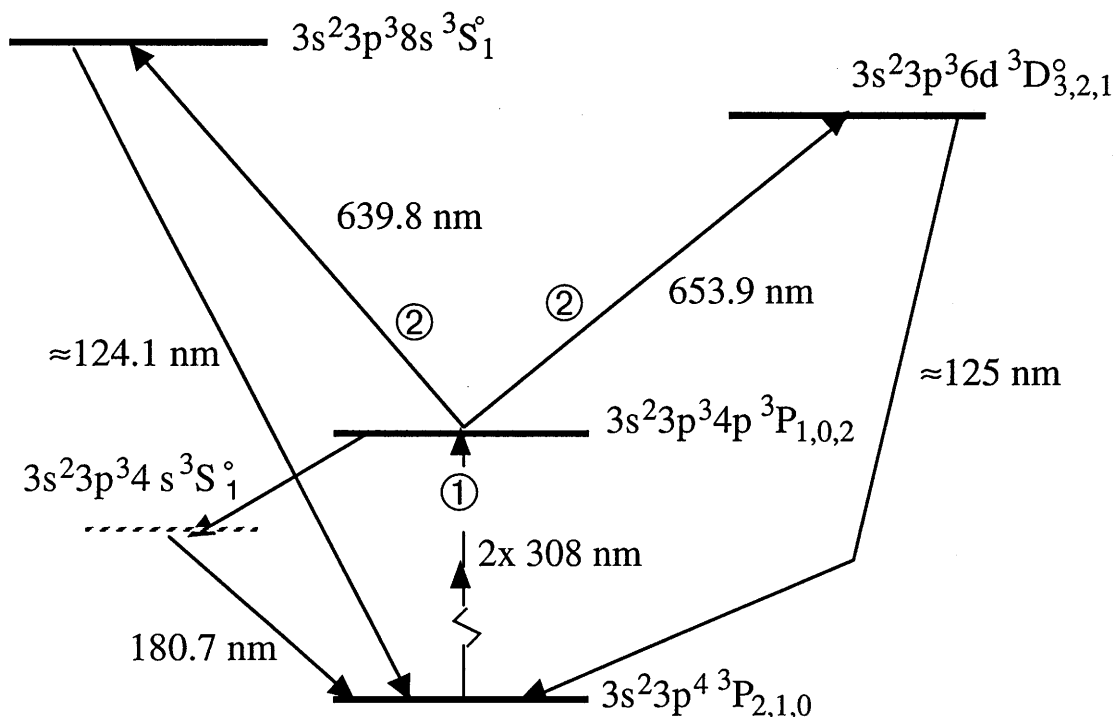


FIG. 1.—S I: partial diagram showing the two-step excitation process

analyzed by computer fitting. The results obtained for the final lifetimes are reported in Table 1. Very similar results were obtained for all fine-structure levels of the 3D term. Thus we give only one lifetime values for all states.

3. LIFETIMES AND ASTRONOMICAL DATA

Since Federman & Cardelli (1995) used the relative strengths for lines within a multiplet given by Morton (1991), the derivation of lifetimes from the astronomical data is straightforward. The multiplet f -values from the *HST* measurements thus are obtained from those compiled by Morton (1991) through a simple scaling; the inferred multiplet values are given in the review by Federman & Cardelli (1996). The multiplet f -value is proportional to the multiplet Einstein A -coefficient and therefore inversely proportional to the lifetime for the upper state. The latter relationship is valid when the decay to the ground state is the primary channel; our HFR results (see § 4) show that this is indeed the case for the multiplets seen in interstellar absorption. The astronomically derived lifetimes are then obtained by taking the inverse of the quantity, $[f_M(\text{FC}) \times A_M(\text{M})]/f_M(\text{M})$, where $f_M(\text{FC})$ is the multiplet f -value given by the analysis of Federman & Cardelli (1995) and $A_M(\text{M})$ and $f_M(\text{M})$ are the multiplet A -Value and f -value listed by Morton (1991). These lifetimes are listed in the sixth column of Table 1. Also shown in the table are other determinations, including the results of the Opacity Project (Butler, Mendoza, & Zeppen 1998).

4. DISCUSSION AND INTERPRETATION

The agreement between the laboratory lifetimes of Berzinsh et al. (1997) and those inferred from the astronomical observations is quite good. The two values are generally very similar, with typical differences within the mutual uncertainties. The astronomical results for the weak lines involving the $6d\ ^3D^{\circ}$ and $8s\ ^3S^{\circ}$ states, however, differ sub-

stantially from the laser measurements obtained in the present work. The comparison with large-scale theoretical calculations is also less satisfactory. In order to discern causes for the differences with the experimental and astronomical results, we performed new computations.

In a previous paper (Berzinsh et al. 1997), an extensive comparison has been made between HFR lifetime values and time-resolved laser spectroscopy measurements. The HFR ab initio predictions made with Cowan's (1981) code were combined with a least-squares fitting of the calculated eigenvalues of the Hamiltonian to the experimental energy levels in order to optimize the Slater parameters. The Slater and configuration interaction integrals, not optimized in the fitting procedure, were scaled down by a factor of 0.85. For the low energy levels, particularly for $4s''\ ^3P^{\circ}$, $4s'\ ^1D^{\circ}$, and $4s\ ^3S^{\circ}$ and, to a lesser extent, for $4p\ ^3P$ and $4p'\ ^3P$ levels, theory was in reasonable agreement with experiment. The discrepancies were found to increase substantially with n , the principal quantum number. Additional separate calculations have been performed in an attempt to explain these discrepancies. These new calculations are briefly summarized here. They are performed in a purely ab initio approach and consequently do not incorporate fitting to the experimental levels; the scaling factors used previously are retained in the calculations.

The effects on the lifetime values of the inclusion of higher members along the Rydberg series and also of the continuum are suspected to play a role. Their eventual contribution was considered through a set of calculations using the following explicit configuration sets: $3s^2 3p^4 + 3s^2 3p^3 np + 3p^6 + 3s 3p^4 3d + 3s 3p^4 4d + 3s 3p^4 4s$ and $3s^2 3p^3 nd + 3s^2 3p^3 ns + 3s 3p^5 + 3s 3p^4 4p$ for the two parities. Different computations were made with the principal quantum numbers chosen lower or equal to $n = 10, 12, 15,$ and $18,$ respectively, this last choice corresponding to the limits of our computer capabilities. Interestingly enough, it was found that most of the lifetime values

TABLE 1
COMPARISON OF EXPERIMENTAL AND THEORETICAL RADIATIVE LIFETIMES^a

LEVEL	ENERGY (cm ⁻¹)	τ_{HFR}		$\tau_{\text{exp}}^{\text{d}}$	FC ^e	OP ^f	τ_{exp} (others)
		This work ^b	Berzinsh ^c				
4s' ³ P ₀ ^o	77136	3.0	2.9			1.9	
4s' ³ P ₁ ^o	77150	2.9	2.9		2.1(0.2)		2.8(0.3) ^g , <1.2 ^h 2.034(0.204) ⁱ
4s' ³ P ₂ ^o	77181	2.8	2.8		2.1(0.2)		2.146(0.256) ⁱ
4s' ¹ D ₂ ^o	69238	1.7	1.6				1.5(0.3) ^j , <1.7 ^h
4s ³ S ₁ ^o	55331	1.5	1.4		1.875(0.188)	1.5	1.5(0.3) ^j , 1.4 ^h 1.875(0.188) ⁱ
5s ³ S ₁ ^o	71351	4.2	3.9	7.1(0.5)	6.9(1.4)	5.5	<6.4 ^h
6s ³ S ₁ ^o	76721	9.7	8.4	17.7(1.1)	16.9(3.4)	14.3	<5 ^h
7s ³ S ₁ ^o	79185	18.8	19.4	35.6(4.0)	31.0(8.4)	30.1	
8s ³ S ₁ ^o	80521	33.1		40(5)*	73.3(44.0)	56.2	
4p ³ P ₀ ^o	64891	36.1	27.3	46.1(1.0)			33(12) ^k
4p ³ P ₁ ^o	64889	36.1	27.3	46.1(1.0)		54.4	33(12) ^k
4p ³ P ₂ ^o	64893	36.0	27.2	46.1(1.0)			33(12) ^k
4p ³ P ₂ ^o	79376	63.0	42.9				53.5(4.0) ^l
5p ³ P ₂ ^o	74269	131.9	103.4	188(13)		209	185(15) ^l
5p ⁵ P ₃ ^o	73921	130.1	103.9			165	615(50) ^l , <97(30) ^k
6p ⁵ P ₃ ^o	77856	289.9	207.9			393	265(20) ^l
7p ⁵ P ₃ ^o	79786	549.8	269.0			729	415(25) ^l
3d ³ D ₁ ^o	70165	1.9	3.4				
3d ³ D ₂ ^o	70166	1.9	3.4		3.0(0.6)	3.0	2.1(0.3) ^j , <1.5 ^h
3d ³ D ₃ ^o	70174	1.9	3.3				
4d ³ D ₁ ^o	75952	6.0	5.7	12.6(1.3)			11.4(1.5) ^g , <8.5 ^h
4d ³ D ₂ ^o	75952	6.0	5.6	12.5(0.7)	9.6(1.9)	13.5	11.4(1.5) ^g , <8.5 ^h
4d ³ D ₃ ^o	75957	5.9	5.5	12.9(1.1)			11.4(1.5) ^g , <8.5 ^h
5d ³ D ₁ ^o	78693	10.1	8.3	41.3			
5d ³ D ₂ ^o	78691	10.0	8.2		37.5(7.5)	61.1	
5d ³ D ₃ ^o	78692	9.9	8.0	40(4)			
6d ³ D ₁ ^o	80186	16.0					
6d ³ D ₂ ^o	80184	15.9		75(7)*	199(90)	227	
6d ³ D ₃ ^o	80182	15.8					
6d ⁵ D ₄ ^o	79992	593.1					260(20) ^l
7d ⁵ D ₄ ^o	80995	994.7	473.				505(40) ^l
8d ³ D ₁ ^o	81668	31.9					
8d ³ D ₂ ^o	81666	31.9			>328	167	
8d ³ D ₃ ^o	81663	31.9					

^a All lifetimes are quoted in nanoseconds. We write $a(b)$ for $a + b$ and b represents a 2σ uncertainty.

^b This work; HFR calculations with Rydberg series limited to $n = 18$ (see the text).

^c Berzinsh et al. (1997); HFR results.

^d Berzinsh et al. (1997) or present work (*), laser spectroscopy.

^e Federman & Cardelli (1995), astronomical sources.

^f Butler et al. (1998); calculations performed in the framework of the Opacity Project.

^g Smith (1978), phase-shift measurement.

^h Müller (1968), wall-stabilized arc.

ⁱ Beideck et al. (1994), beam-foil spectroscopy.

^j Savage & Lawrence (1966), phase-shift technique.

^k Bridges & Wiese (1968), arc measurements.

^l Delalic et al. (1990), high-frequency-deflection technique.

increase with n when they are compared to the previous HFR results (Berzinsh et al. 1997). The only exceptions are $4s'^1D_2^o$ for which the τ -value remains remarkably constant but in agreement with the measurements of Savage & Lawrence (1966) and of Müller (1968) and the $3d^3D_{1,2,3}^o$ levels the lifetimes of which are somewhat decreased but are now again in better agreement with the measurements of Savage & Lawrence (1966). In a general way, the agreement between the new results and the laser measurements (Berzinsh et al. 1997) is improved, the improvement being strikingly pronounced in the case of the np^3P ($n = 4-7$) terms. In the quintet system of the np series, the new HFR lifetime for $6p^5P_3$ is now in good agreement with the result reported by Delalic, Erman, & Källne (1990) while it is higher for $7p^5P_3$. Some convergence is also observed in the process of lifetime calculation. The results obtained by limiting the series to $n = 15$ are close to those obtained with

the additional configurations up to $n = 18$, but marginal differences still appear for the nd^3D^o series. From the above considerations, it can be inferred that part of the remaining discrepancies between theory and experiment could probably be decreased by using more extensive configuration basis sets (including the continuum), but this would require much more powerful computers. It is worth noting that the lifetimes from the Opacity Project (Butler et al. 1998) tend to be larger than our predictions, with the empirical results lying between the two sets of computed lifetimes. In a general way, the calculations of the Opacity Project have been performed using the close-coupling approximation of electron-atom collision theory. The target functions were constructed using conventional methods of configuration interaction theory and the radial functions were calculated using either SUPERSTRUCTURE or CIV3 codes. The details have been given by Berrington et al. (1987). It should

TABLE 2
TRANSITION PROBABILITIES (*A*) AND OSCILLATOR STRENGTHS (*f*) FOR VUV LINES OF ASTROPHYSICAL INTEREST^a

UPPER LEVEL	LOWER LEVEL	λ_{exp}^b (nm)	THIS WORK			PREVIOUS (<i>f</i> -value)		BR ^g (%)	
			A (s ⁻¹) ^c	^d	<i>f</i> -value	^e	^f		
5s ³ S ₁ ^o	3p ⁴ ³ P ₁	140.9337	7.057(7)	4.194(7)	1.294(-2)		1.489(-2)	29.78	
		³ P ₀	141.2873	2.307(7)	1.371(7)	1.273(-2)		1.436(-2)	9.74
		³ P ₂	140.1514	1.216(8)	7.225(7)	1.321(-2)	1.49(-2)	1.608(-2)	51.31
6s ³ S ₁ ^o	3p ⁴ ³ P ₁	131.0194	2.983(7)	1.639(7)	4.364(-3)		2.894(-2)	29.02	
		³ P ₀	131.3249	9.743(6)	5.355(6)	4.295(-3)		2.887(-2)	9.48
		³ P ₂	130.3430	5.190(7)	2.852(7)	4.508(-3)	5.06(-3)	2.909(-2)	50.49
7s ³ S ₁ ^o	3p ⁴ ³ P ₁	126.9209	1.495(7)	7.912(6)	1.976(-3)			28.17	
		³ P ₀	127.2075	4.783(6)	2.532(6)	1.905(-3)			9.01
		³ P ₂	126.2860	2.696(7)	1.427(7)	2.119(-3)	2.59(-3)		50.79
8s ³ S ₁ ^o	3p ⁴ ³ P ₁	124.8045	8.980(6)	7.464(6)	1.758(-3)			29.71	
		³ P ₀	125.0814	3.100(6)	2.577(6)	1.875(-3)			10.26
		³ P ₂	124.1905	1.421(7)	1.181(7)	1.695(-3)	1.06(-3)		47.03
4d ³ D ₁ ^o	3p ⁴ ³ P ₁	132.3522 ^h	6.933(7)	3.300(7)	8.930(-3)		1.020(-2)	41.60	
		³ P ₀	132.6643	9.067(7)	4.316(7)	3.516(-2)		4.072(-2)	54.39
		³ P ₂	131.6618 ^h	4.827(6)	2.298(6)	3.690(-4)	4.54(-4)	4.106(-4)	2.90
4d ³ D ₂ ^o	3p ⁴ ³ P ₁	132.3516 ^h	1.231(8)	5.869(7)	2.642(-2)		3.061(-2)	73.36	
		³ P ₂	131.6618 ^h	4.282(7)	2.041(7)	5.458(-3)	6.79(-3)	6.154(-3)	25.51
		¹ D ₂	149.8942	2.962(4)	1.412(4)	5.120(-6)			0.02
4d ³ D ₃ ^o	3p ⁴ ³ P ₂	131.6542	1.681(8)	7.667(7)	2.871(-2)		3.81(-2)	3.447(-2)	99.05
		¹ D ₂	149.8850	3.046(4)	1.389(4)	7.100(-6)			0.02
		¹ D ₃	149.8850	3.046(4)	1.389(4)	7.100(-6)			0.02
5d ³ D ₁ ^o	3p ⁴ ³ P ₁	127.7199	4.167(7)	1.024(7)	2.570(-3)		1.671(-2)	41.97	
		³ P ₀	128.0099	5.340(7)	1.312(7)	9.931(-3)		6.607(-2)	53.79
		³ P ₂	127.0769 ^h	3.013(6)	7.402(5)	1.104(-4)	1.09(-4)	6.623(-4)	3.03
5d ³ D ₂ ^o	3p ⁴ ³ P ₁	127.7212	7.228(7)	1.787(7)	7.480(-3)		4.930(-2)	72.38	
		³ P ₂	127.0782 ^h	2.636(7)	6.518(6)	1.622(-3)	1.64(-3)	1.002(-2)	26.40
		¹ D ₂	143.9828 ^h	1.686(4)	4.170(3)	1.390(-6)			0.02
5d ³ D ₃ ^o	3p ⁴ ³ P ₂	127.0782 ^h	9.991(7)	2.470(7)	8.611(-3)		9.02(-3)	5.508(-2)	98.80
		¹ D ₂	143.9817 ^h	1.894(4)	4.683(3)	2.190(-6)			0.02
		¹ D ₃	143.9817 ^h	1.894(4)	4.683(3)	2.190(-6)			0.02
6d ³ D ₁ ^o	3p ⁴ ³ P ₁	125.3297	2.405(7)	5.133(6)	1.245(-3)		9.613(-3)	38.39	
		³ P ₀	125.6093	3.640(7)	7.770(6)	5.665(-3)		3.802(-2)	58.12
		³ P ₂	124.7107 ^h	1.224(6)	2.613(5)	3.760(-5)	1.92(-5)	3.811(-4)	1.95
6d ³ D ₂ ^o	3p ⁴ ³ P ₁	125.3325	4.916(7)	1.042(7)	4.208(-3)		2.903(-2)	77.98	
		³ P ₂	124.7134 ^h	1.291(7)	2.738(6)	6.565(-4)	2.90(-4)	5.768(-3)	20.48
		¹ D ₂	140.9538 ^h	6.068(3)	1.287(3)	4.114(-7)			0.01
6d ³ D ₃ ^o	3p ⁴ ³ P ₂	124.7160	6.240(7)	1.316(7)	4.428(-3)		1.64(-3)	3.244(-2)	98.46
		¹ D ₂	140.957 ^h	1.302(4)	2.747(3)	1.228(-6)			0.02

^a As deduced from the laser measurements and HFR branching ratios. We write $a(-b)$ for $a \times 10^{-b}$.

^b From Kaufman & Martin 1993 or, otherwise, calculated from the levels given by Martin et al. 1990, unless otherwise noted.

^c This work: HFR theoretical values.

^d Calculated from the laser spectroscopy lifetimes (Berzinsh et al. 1997 and present work) and the HFR branching ratios.

^e Federman & Cardelli 1995.

^f Morton 1991.

^g This work: HFR branching ratios.

^h Blend.

be emphasized that the fine structure is not considered within the framework of the Opacity Project calculations. Consequently, only multiplet transition probabilities are provided while the HFR method supplies data for individual lines.

An additional separate calculation was performed in order to estimate the importance of additional selected configurations within the Layzer complexes $n = 4$ and $n = 5$ not included in the basis used by Berzinsh et al. (1997). More specifically we have added the configurations $3p^2 4s^2$, $3p^2 4p^2$, $3p^2 3d^2$, $3p^2 4d^2$, $3p^2 5p^2$, $3p^2 5d^2$, $3p^2 4s 4d$, $3p^2 4s 5d$, and $3p^3 4s^2 4p$ and $3p^2 4s 4p$, $3p^2 4s 5p$, $3p^3 4s^2 3d$, $3p^3 4s^2 4d$, and $3p^3 4s^2 5d$, respectively, to the configuration sets used previously (limited to $n = 5$). The dimensions of the matrices obtained and the limits imposed by Cowan's code (no more than eight open subshells) have prevented us from performing a more detailed analysis of configuration interaction effects. However, the increase in the lifetimes obtained for the first two members of the series was sub-

stantial. The new values for $4p^3 P_{0,1,2}$ ($\tau = 39.7, 39.6,$ and 39.5 ns) and $5p^3 P_{0,1,2}$ ($\tau = 168.4, 168.3, 167.6$ ns) levels are now in good agreement with the laser lifetimes measured at the Lund Laser Center. On the other hand, the effects on the $nd^3 D$ series are much smaller, with the changes induced for $n = 5$ reaching only a few (less than 5 percent).

Finally, the effect on the theoretical lifetimes of opening the inner $2p$ subshell was investigated *qualitatively*. Inner shell excitations could eventually modify the oscillator strengths of some transitions. This has been pointed out in another context in the case of the iron group elements (see Quintet & Hansen 1995). Thus the effects of configurations $2p^5 3s^2 3p^5$, $2p^5 3s^2 3p^4 4p$, and $2p^5 3p^6 4p$ for the even parity and $2p^5 3s^2 3p^4 3d$, $2p^5 3s^2 3p^4 4d$, $2p^5 3s^2 3p^4 4s$, $2p^5 3p^6 4s$, $2p^5 3p^6 3d$, and $2p^5 3p^6 4d$ for the odd parity were not able to explain the remaining discrepancies between theory and experiment; the lifetimes generally *decreased* with the inclusion of such configurations. However, these results are only qualitative. A more quantitative estimation would require

the simultaneous consideration of much more correlation in the atomic model, which is not possible with our computer capabilities.

By combining the laser lifetime measurements and the theoretical branching ratios, it is possible to derive a consistent set of transition probabilities and f -values for the UV transitions of astrophysical interest. The results are reported in Table 2 where they are compared with the astrophysical results deduced from the astronomical (ISM) spectra by Federman & Cardelli (1995) and with the results taken from Morton's (1991) compilation. The agreement is excellent when comparing the present results with the ISM values deduced by Federman & Cardelli (1995), except for the relatively weak transitions involving the $6d^3D^o$ and $8s^3S^o$ levels where the discrepancies reflect the differences

observed in the lifetimes. Large discrepancies are observed for some transitions, i.e., those involving $6s^3S^o$, $5d^3D^o$, and $6d^3D^o$, when comparing the present results with the f -values taken from the compilation of Morton (1991), as noted earlier by Federman & Cardelli (1995). In light of the agreement among the empirical measures, the f -values reported here can be used with confidence in future astronomical studies.

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