

## Lifetimes of singly excited states in He I

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We report a comprehensive set of calculations of the Rydberg-state lifetimes in the helium atom, using a realistic potential to represent the atomic core. The effects due to blackbody radiation are explicitly included. The overall results compare well with experiment for all principal quantum numbers examined and should provide guidance for future experimental investigations. They are used here to single out incorrect experimental determinations of lifetimes. Our calculated oscillator strengths are in excellent agreement with predictions of previous and very sophisticated calculations.

### I. INTRODUCTION

Being the simplest nontrivial atom, helium has traditionally been the subject of many experimental studies and the testing ground for the great majority of theoretical approaches to various atomic processes. Very recently, laser spectroscopy has established extremely accurate energy splittings between Rydberg states.<sup>1,2</sup> Availability of such data has made possible the semiempirical parametrization of Rydberg series and accurate predictions of higher-lying states<sup>3</sup> for which experimental data are not available or hard to obtain. It appears that predictions of energy intervals by such methods,<sup>3</sup> and in some cases by *ab initio* calculations,<sup>4</sup> are of comparable accuracy with experiment.

In experimental investigations of the high-lying Rydberg states, especially of their photoexcitation and their collisional excitation, knowledge of their lifetimes and various transition probabilities to and from them is needed (a) in assessing the importance of the various processes taking place, and (b) in the direct determination of other quantities such as the "optical" cross sections for electron excitation measured by Jobe and St. John.<sup>5</sup>

Accurate theoretical calculation of transition probabilities is usually a difficult task and, in most cases, the accuracy is considerably inferior to that of the experimental values. Theoretical works normally produce values of absorption oscillator strengths and line strengths. There is a plethora of such calculations for helium with varying degrees of sophistication and accuracy; Crossley<sup>6</sup> has reviewed and compared the more important ones, for transitions between the low-lying states. The most representative works are the following. (a) The very accurate variational calculations using many-term expansions in terms of Hylleraas-type wave functions. This approach has been widely employed by Pekeris and co-workers,<sup>7</sup> by Anderson and Weinhold,<sup>8</sup> who established rigorous upper and lower bounds for the oscillator-strength values, by Scherr and co-workers,<sup>9</sup> and by Weiss.<sup>10</sup> (b) Central-field calculations including effects of electron exchange and configuration interaction<sup>11</sup> (CI). (c) Hartree-Fock calculations with various approaches of treating electron correlations,<sup>12</sup> especially the work of Trefftz and co-workers,<sup>12(b)</sup> who evaluated the electron correlation by introducing the

factor  $1+ar_{12}$  in the two-electron wave function. (d) Estimates using the Coulomb approximation developed by Bates and Damgaard.<sup>13</sup>

All the calculations, except under class (a) above, use the experimentally available energy values. Because of their considerably involved nature, calculations of the types (a)–(c) have usually been limited to the lowest few states. The Hylleraas-type calculations demand an ever-increasing number of basis functions to correctly describe states with increasing principal quantum number  $n$ . This is also true for the configuration-interaction calculations. The nature of these two approaches implies<sup>7</sup> that the former are relatively more accurate for the lower states, where electron correlations are the strongest, and the latter are best for higher states, where correlations are less important. Very precise theoretical transition probabilities in He exist for only a few transitions. The most extensive oscillator strengths available are by Green *et al.*<sup>11</sup> within the CI approximation and for transitions between the series  $^1S-^1P$ ,  $^3S-^3P$ ,  $^1P-^1D$ , and  $^3P-^3D$ , for principal quantum numbers  $n \leq 9$ . Gabriel and Heddle<sup>14</sup> tabulated the transition probabilities corresponding to these states, as calculated by Dalgarno and co-workers. Values for higher principal quantum numbers are not systematically available and estimation of the relevant state lifetimes is done by some kind of extrapolation, or by the Coulomb approximation.<sup>13</sup> The compilation of data by Wiese *et al.*<sup>15</sup> has been widely used in the literature, but this work is incomplete, dated, and has no data for  $n > 12$ . There are no published transition probabilities for *d-f* and *f-g* transitions, although they are expected to be closely approximated by the relevant hydrogenic values.

The lifetime of a state depends on the transition probabilities from this state to all dipole-accessible states, in the radiative decay case. Therefore, it is dependent on the properties of several other states, and an accurate description of them *all* is necessary for the accurate theoretical prediction of the lifetime. In addition, a reasonable estimation of cascade contributions from higher states is usually necessary for the experimental determination of a lifetime. Recent measurements at low  $n$  by Bukow *et al.*<sup>16</sup> and by Thompson and Fowler<sup>17</sup> indicated only fair agreement with calculated lifetimes, whereas some of the also experimental values of Hitashi *et al.*<sup>18</sup> differ

from the theoretical ones by a factor of 2 for some medium  $n$  values. Finally, the experimental energy values, normally used as input in theoretical calculations, have also been revised by more recent measurements; the latest compilation of the new frequency values is provided by Martin.<sup>19</sup>

In view of the above situation, the present systematic study was undertaken of the transition probabilities and oscillator strengths between all  $s$ ,  $p$ ,  $d$ ,  $f$ , and  $g$  states up to  $n=21$ . Our modest objective was to provide values of these quantities with a 10% accuracy, in order to assist experimentalists in estimating the lifetimes of excited states and the importance of relevant processes. To this end we employed an approach developed by this author which yielded accurate predictions of lifetimes in the alkali-metal atoms.<sup>20</sup> We also explicitly accounted for the effects of blackbody radiation present in practically all experimental investigations. Our method is a numerical extension of the Coulomb approximation, using a central-field potential to represent the atomic core; when appropriate, this potential is augmented by polarization and spin-orbit interaction terms.<sup>20</sup> In Sec. II we give only a brief outline of the method of calculation.

## II. PRESENT APPROACH

The absorption oscillator strength and the transition probability between two levels  $|\gamma J\rangle$  and  $|\gamma' J'\rangle$  are given by

$$f(\gamma' J' \rightarrow \gamma J) = \frac{1}{3} \frac{\Delta E}{R} \frac{|(\gamma S L J || \vec{e} \cdot \vec{r} || \gamma' S' L' J')|^2}{2J'+1}, \quad (1)$$

and

$$A(\gamma J \rightarrow \gamma' J') = 4\alpha^2 \left[ \frac{\Delta E}{R} \right]^2 \frac{f(\gamma' J' \rightarrow \gamma J)}{2J+1}, \quad (2)$$

respectively, where  $\alpha$  is the fine-structure constant,  $R$  is the reduced-mass Rydberg energy constant, and  $\Delta E = E(\gamma J) - E(\gamma' J')$  is the transition energy. For the transitions considered here the energy splittings within the triplet multiplets are small and experimental measurements of lifetimes do not differentiate between them. Therefore, we calculated the total oscillator strengths and transition probabilities. In this case, the square of the reduced matrix element takes the simple form

$$|(1s n l; S L || r || 1s n' l'; S' L')|^2 = (2S+1) \max(l, l') [R(n l, n' l')]^2, \quad (3)$$

$$R(n l, n' l') = \int_0^\infty dr P_{n l}(r) r P_{n' l'}(r). \quad (4)$$

The electronic wave functions  $P_{n l}(r)$ , being central to this work, were obtained by *inward* integration of the Schrödinger equation

$$d^2 P_{n l}(r) / dr^2 - [V(r) - E_{n l} - l(l+1)/r^2] P_{n l}(r) = 0, \quad (5)$$

where  $V(r)$  is the potential experienced by the active electron and  $E_{n l}$  is its eigenvalue. We employed values for  $E_{n l}$  deduced from experimental (spectroscopic) energies,<sup>14</sup>

eliminating the need of a self-consistent calculation. The atomic potential was taken to be the Hartree-Slater prediction<sup>21</sup> for the ground state of neutral helium and was kept the same for all states. The wave function  $P_{n l}(r)$  in Eq. (5) is well defined and exactly known at large distances  $r$ , being the Whittaker function. The traditional Coulomb approximation is based on the realization of this fact and on the *assumption* that the atomic field is equal to its asymptotic value,  $\sim 1/r$ , for *all* distances. The matrix element (4) is then evaluated analytically. The error due to the above assumption about small distances is (partially) corrected for by a suitable choice of an integration cut-off distance.<sup>13</sup>

In our treatment the wave function is obtained by *inward numerical* integration of Eq. (5) with the full atomic potential  $V(r)$ . The integration starts from practical infinity and with the (correct) asymptotic boundary condition (a Whittaker function). The integration is continued until the inner classical turning point (very close to the origin). The inward solution is then matched to the outward solution, from the origin, and no iteration is performed. This approach ensures minimal propagation of the error due to *any* degree of incorrectness of the atomic potential used. The possible discontinuity of the wave function's first derivative at the matching point is inconsequential to the calculation of the transition matrix elements (3).

As already mentioned above, this approach was successfully applied to the alkali-metal atoms.<sup>20</sup> In principle, it is applicable to any other case, even when strong interactions between Rydberg series are present, provided the interactions are included into the full wave function via an expansion in terms of *LS*-coupling wave functions. Coefficients of such an expansion could be obtained by, e.g., the semiempirical analysis of available experimental levels.<sup>22</sup>

## III. RESULTS AND DISCUSSION

Using the above approach, we obtained the wave functions of the relevant  $s$ ,  $p$ ,  $d$ ,  $f$ , and  $g$  states with principal quantum number up to  $n=21$ , and the calculated matrix elements were used in Eqs. (1) and (2) to yield the transition probabilities and oscillator strengths between all these states. Since a number of calculations were available<sup>7-12</sup> for the low- $n$  states, we tested the accuracy of our predictions. Table I gives the comparison between our values and the ones considered the most accurate and rigorous in the literature.<sup>7,8,10</sup> The agreement is impressive. One of the reasons for such an agreement is attributed to the use of experimental energy values. We should recall, however, that this is not an artificial treatment, since all calculations, other than the Hylleraas-type, also use experimental frequencies. One is generally fortunate in the case of helium in that no doubly excited states exist below the first ionization threshold. Thus the considerable added complications due to transitions to and from such states are absent. The seemingly lower values obtained by the present approach for higher  $n$ , compared to the results of Refs. 7 and 8, may be due to two plausible reasons: (i) the

TABLE I. Comparison of calculated oscillator strengths of the present work with those of Schiff *et al.* (Ref. 7), Green *et al.* (Ref. 11), and Anderson and Weinhold (Ref. 8).

Transition	This work	Schiff <i>et al.</i>	Green <i>et al.</i>		Anderson and Weinhold
			<i>L</i>	<i>V</i>	
$1^1S-2^1P$	0.2761	0.2762	0.275 37	0.275 86	0.2761(14)
$1^1S-3^1P$	0.073 34	0.733	0.072 92	0.072 96	0.0735(36)
$1^1S-4^1P$	0.029 81	0.030	0.029 57	0.029 60	0.0303(71)
$1^1S-5^1P$	0.015 00	0.015	0.014 81	0.014 87	
$2^1S-2^1P$	0.3754	0.3764	0.3773	0.3950	0.3764(18)
$2^1S-3^1P$	0.1489	0.1514	0.1513	0.1540	0.151(11)
$2^1S-4^1P$	0.0483	0.049	0.0493	0.0506	0.052(18)
$2^1S-5^1P$	0.0220	0.02	0.0224	0.0231	
$3^1S-2^1P$	0.1438	0.1454	0.1457	0.1446	0.1454(91)
$3^1S-3^1P$	0.6257	0.626	0.6279	0.6448	0.626(11)
$3^1S-4^1P$	0.1433	0.144	0.1429	0.1433	0.148(45)
$3^1S-5^1P$	0.0502	0.05	0.0499	0.0506	
$4^1S-2^1P$	0.0255	0.0258	0.0260	0.0256	
$4^1S-3^1P$	0.3070	0.306	0.3081	0.3092	
$4^1S-4^1P$	0.8578	0.85	0.8603	0.8754	
$4^1S-5^1P$	0.1460	0.15	0.1455	0.1445	
$5^1S-2^1P$	0.009 49	0.0096	0.009 67	0.009 51	
$5^1S-3^1P$	0.0553	0.055	0.0554	0.0556	
$5^1S-4^1P$	0.4756	0.47	0.4767	0.4780	
$5^1S-5^1P$	1.0833	1.1	1.0869	1.0901	
$2^3S-2^3P$	0.537 532	0.539 086	0.5398	0.5487	0.539 09(47)
$2^3S-3^3P$	0.062 05	0.064 46	0.0644	0.0668	0.0645(29)
$2^3S-4^3P$	0.024 81	0.025 77	0.0259	0.0271	0.0261(63)
$2^3S-5^3P$	0.0120	0.0125	0.0125	0.0132	
$3^3S-2^3P$	0.204 45	0.208 54	0.2087	0.2055	
$3^3S-3^3P$	0.8884	0.8909	0.8922	0.9110	
$3^3S-4^3P$	0.0497	0.0501	0.0499	0.0491	
$3^3S-5^3P$	0.0227	0.023	0.0228	0.0226	
$4^3S-2^3P$	0.0310	0.0317	0.0318	0.0312	
$4^3S-3^3P$	0.4339	0.4357	0.4363	0.4384	
$4^3S-4^3P$	1.2136	0.2153	1.2164	1.2305	
$4^3S-5^3P$	0.0441	0.044	0.0439	0.0430	
$5^3S-2^3P$	0.0111	0.0113	0.0113	0.0111	
$5^3S-3^3P$	0.0673	0.068	0.0693	0.0679	
$5^3S-4^3P$	0.6673	0.67	0.6605	0.6742	
$5^3S-5^3P$	1.5295	1.53	1.5369	1.5383	

use of a "frozen" core to describe the potential for the excited states; or (ii) the increased difficulties of the Hylleraas-type function expansions for higher states. We feel that our oscillator strengths are accurate and they will be tabulated in detail elsewhere<sup>23</sup> for all the transitions between the states with  $n \leq 21$  and  $l \leq 5$ .

The calculated transition probabilities were used to obtain the lifetimes of the singly excited states  $1snl^{1,3}L$  of He and for  $L=S-F$ . The results are given in Table II. The lifetimes shown there depend on the transition probabilities to all dipole-allowed lower states, and the compos-

ite final result is affected to various degrees by the inadequacies of the approximation used, depending on the upper state under consideration. When comparing with experiment, we must take into account the experimental conditions, and especially the temperature at which the measurements were performed. The population of the higher states can be significantly depleted by stimulated emission and absorption, due to the blackbody radiation in which the atoms are embedded.<sup>24,25,20</sup> The total transition probability  $A_m^*$  from the  $m$ th state to "anywhere" is generally obtained<sup>20</sup> from

TABLE II. Lifetimes of (a)  $1snl^1L$  and (b)  $1snl^3L$  states in He (in nsec). Capital letters denote references in part (c).

State	0 K	This work		(a)	Experimental	Other theoretical	
		300 K	600 K				
$3^1S$	55.275	55.272	55.180	50.3(2.3) 55.9(2) 60(3) 55(6) 54.1(6)	BHR TF OV BKM KB	55.2(1.7)	WSG
$4^1S$	88.865	88.761	88.363	77.9(3.5) 89(1) 88(1) 75(1) 87(1.5) 89(3) 75(4) 86.2(2.5) 97(2) 84(9) 87(1.5)	BHR TF KOSB IR CBHG JF OV BK2 PH BKM KB	89.8(4.0) 89.8	WSG GH
$5^1S$	147.93	147.93	147.61	109(16) 160(3) 145(6) 118(8) 115(5) 152(5) 144(3) 141 133(18)	BHR TF KOSB AJS OV BK2 PH BKM KB	150(12) 151	WSG GH
$6^1S$	234.95	233.71	229.51	230(7) 210(4) 204(6) 235(8)	LMP TF HDKKD KOSB	233 240(12) 240.4	GJK WSG GH
$7^1S$	354.69	351.08	339.93	360(18) 350(100)	LMP TF	351 356(35) 356	GJK WSG GH
$8^1S$	512.93	503.63	480.67	513(30) 450(150) 283(40)	LMP TF HDKKD	508 520(52) 520	GJK WSG GH
$9^1S$	712.76	692.73	652.07	625(40)	LMP	710	GJK
$10^1S$	962.19	924.41	858.73				
$11^1S$	1265.00	1200.39	1101.29				
$12^1S$	1626.08	1523.42	1381.38				
$13^1S$	2057.72	1902.92	1706.31				
$14^1S$	2557.11	2334.28	2071.23				
$15^1S$	3131.59	2822.43	2479.73				
$16^1S$	3774.96	3360.43	2925.49				
$17^1S$	4513.93	3970.72	3427.16				
$18^1S$	5344.49	4649.01	3981.10				
$19^1S$	6272.04	5401.79	4594.20				
$20^1S$	7301.91	6239.07	5278.12				
$21^1S$	8427.70	7171.85	6051.04				
$2^1P$	0.5548	0.5548	0.5548				
$3^1P$	1.7272	1.7272	1.7272	1.7225(46) 1.70(4) 1.74(2) 1.72(10) 1.80(10) 1.73(11) 1.78(10)	ACLMM BHR ACLMS KKC CDDG BL MB	1.726(2) 1.725(17) 1.74 1.73 1.73 1.72(9) 1.76	SPA WSG GJK W DS AW CME

TABLE II. (Continued).

State	0 K	This work 300 K	600 K	Experimental		Other theoretical	
4 <sup>1</sup> P	3.974	3.973	3.973	4.05(12) 4.02(10) 3.7(4)	BHR ACLMS MB	3.92(11)	WSG
5 <sup>1</sup> P	7.651	7.650	7.647				
6 <sup>1</sup> P	13.10	13.10	13.08				
7 <sup>1</sup> P	20.69	20.68	20.63	22.7(3.4)	LMP	21 20.4	GJK GH
8 <sup>1</sup> P	30.76	30.72	30.63	31.7(4.0)	LMP	31.1 30.5	GJK GH
9 <sup>1</sup> P	43.66	43.59	43.41	43.6(3.1)	LMP	40	LMP
10 <sup>1</sup> P	59.81	59.66	59.36	54.9(3.8)	LMP	52	LMP
11 <sup>1</sup> P	79.46	79.20	78.74	71.4(7.1)	LMP	73	LMP
12 <sup>1</sup> P	103.02	102.60	101.93	90.9(9.1)	LMP	91	LMP
13 <sup>1</sup> P	130.86	130.22	129.28	125(20)	LMP		
14 <sup>1</sup> P	163.29	162.37	161.09				
15 <sup>1</sup> P	200.70	199.42	197.75				
16 <sup>1</sup> P	243.42	241.72	239.57				
17 <sup>1</sup> P	291.80	289.62	286.96				
18 <sup>1</sup> P	346.33	343.62	340.41				
19 <sup>1</sup> P	406.85	403.69	400.00				
20 <sup>1</sup> P	474.41	471.10	467.24				
21 <sup>1</sup> P	547.33	545.29	542.67				
3 <sup>1</sup> D	15.69	15.69	15.69	15.2(5) 20(1) 17(2) 15.8(1) 20.3(3) 13.4(5) 15.5(5) 20.5(9) 12(3) 16(1) 16(2) 18(5) 16(4) 16.5(1) 16.5(2)	BHR TF KKC B CBD IR AJS DW D OV PH FHJC KB FNPP DPPB	15.67(47)	WSG
4 <sup>1</sup> D	36.96	36.96	36.95	32.9(2.3) 41(3) 33(7) 38.4(2.1) 39.2(8) 33.6(3) 41(3) 38(3) 34(1) 38(5) 38(2) 41(5) 30(2) 39(5) 47(5) 35(4) 38(1) 30(5) 39.1(2.0)	BHR TF KKC BJ B CBD JF MBBBLBB CBHG AJS DW D OV BK1 PH FHJC FNPP KB DPPB	36.6(1.2)	WSG

TABLE II. (Continued).

State	0 K	This work 300 K	600 K	Experimental		Other theoretical	
5 <sup>1</sup> D	71.52	71.52	71.25	63.5(5.7)	BHR	72.0(2.3)	WSG
				56(10)	TF		
				52(6)	YHM		
				80(40)	KKC		
				74.4(5)	CBD		
				71.9(1.8)	B		
				66(4)	MBBBLBB		
				46(3)	AJS		
				68.0(7.0)	DW		
				49(5)	D		
				49(5)	OV		
				63(9)	BK1		
				79(6)	PH		
				46(3)	KB		
				49.1(2.0)	DPPB		
6 <sup>1</sup> D	122.57	122.43	121.03	72(3)	TF		
7 <sup>1</sup> D	193.29	192.49	188.47				
8 <sup>1</sup> D	286.83	284.25	275.74				
9 <sup>1</sup> D	406.74	400.59	385.24				
10 <sup>1</sup> D	555.85	543.66	518.69				
11 <sup>1</sup> D	737.50	716.03	678.11				
12 <sup>1</sup> D	954.92	920.12	865.37				
13 <sup>1</sup> D	1213.74	1160.83	1085.15				
14 <sup>1</sup> D	1511.79	1435.70	1334.96				
15 <sup>1</sup> D	1855.19	1750.21	1619.73				
16 <sup>1</sup> D	2250.83	2110.66	1945.15				
17 <sup>1</sup> D	2693.44	2513.23	2308.96				
18 <sup>1</sup> D	3186.48	2963.21	2717.66				
19 <sup>1</sup> D	3751.98	3484.44	3195.18				
20 <sup>1</sup> D	4372.39	4076.16	3753.74				
21 <sup>1</sup> D	5049.52	4793.77	4488.63				
4 <sup>1</sup> F	72.31	72.31	72.23	80(6)	BHR	72.5(7.2)	WSG
5 <sup>1</sup> F	139.74	139.69	138.34				
6 <sup>1</sup> F	238.81	238.10	231.43				
7 <sup>1</sup> F	375.50	371.79	353.90	400(50)	TF	370	TF
8 <sup>1</sup> F	555.95	544.52	508.74				
9 <sup>1</sup> F	786.14	760.00	698.68	750(100)	TF	750	TF
10 <sup>1</sup> F	1072.40	1022.24	926.62	1100(200)	TF	1100	TF
11 <sup>1</sup> F	1420.39	1334.40	1194.57	1350(300)	TF	1270	TF
12 <sup>1</sup> F	1836.39	1700.19	1504.96				
13 <sup>1</sup> F	2326.74	2124.39	1863.19				
14 <sup>1</sup> F	2897.52	2611.29	2272.05				
15 <sup>1</sup> F	3554.68	3165.09	2734.90				
16 <sup>1</sup> F	4304.39	3792.18	3258.19				
17 <sup>1</sup> F	5152.88	4500.87	3851.11				
18 <sup>1</sup> F	6106.12	5301.71	4526.01				
19 <sup>1</sup> F	7170.17	6216.99	5312.51				
20 <sup>1</sup> F	8351.28	7301.56	6288.91				
21 <sup>1</sup> F	9642.06	8740.85	7750.56				
(b)							
3 <sup>3</sup> S	36.52	36.52	36.50	35.1(1.3)	BHR	36.0(1.1)	WSG
				57(1)	TF		
				47(3)	OV		
				40.8(8)	KB		

TABLE II. (Continued).

State	0 K	This work 300 K	600 K	Experimental		Other theoretical	
4 <sup>3</sup> S	63.31	63.27	62.97	62.0(2.7)	BHR	58.4(4.2)	WSG
				62(3)	TF	64(6)	TF
				59.2(6)	KOSB		
				63.5(1.2)	IR		
				69(3)	OV		
				62.9(4.4)	BK2		
				68(1)	PH		
				64.5(4.0)	BKM		
				59(6)	FHJC		
				77.5(4.0)	BD		
				67.5(1.0)	HWR		
5 <sup>3</sup> S	110.14	109.82	108.70	100(15)	BHR	111.0(6.7)	WSG
				120(20)	TF	110.4	GH
				111(1)	KOSB		
				106(5)	OV		
				115(6)	BK2		
				113(4)	PH		
6 <sup>3</sup> S	180.05	178.86	175.21	179(4)	KOSB	183.2	GH
7 <sup>3</sup> S	277.37	273.84	264.72	260(10)	KOSB	260	GH
8 <sup>3</sup> S	406.60	397.89	379.68	368(7)	KOSB	372	GH
9 <sup>3</sup> S	572.44	554.37	522.40	390(50)	HDKKD	500	FI
10 <sup>3</sup> S	779.51	746.44	694.98	450(100)	HDKKD		
11 <sup>3</sup> S	1032.48	976.89	898.97				
12 <sup>3</sup> S	1335.24	1247.76	1135.28				
13 <sup>3</sup> S	1694.61	1563.89	1407.53				
14 <sup>3</sup> S	2113.18	1925.63	1714.94				
15 <sup>3</sup> S	2595.15	2334.99	2058.48				
16 <sup>3</sup> S	3155.89	2805.06	2449.32				
17 <sup>3</sup> S	3783.12	3320.28	2871.63				
18 <sup>3</sup> S	4485.23	3887.17	3330.91				
19 <sup>3</sup> S	5275.29	4515.85	3835.59				
20 <sup>3</sup> S	6153.50	5203.49	4381.78				
21 <sup>3</sup> S	7119.62	5949.02	4968.73				
22 <sup>3</sup> S	7985.21	6611.61	5488.61				
2 <sup>3</sup> P	98.17	98.17	98.17				
3 <sup>3</sup> P	98.11	98.10	98.00	89(5)	BHR	94.71(90)	BHR
				122(5)	TF	96.6(1.0)	WSG
				89(10)	S		
				98(4.3)	B		
				111(5)	CBHG		
				122(5)	JF		
				93(12)	MBBBLBB		
				89(5)	D		
				115(5)	OV		
				100(3)	BK1		
				115(2)	PH		
				91(8)	FHJC		
				95.8(6.0)	DPPB		
				106(5)	BD		
				115(5)	HWR		
4 <sup>3</sup> P	142.83	142.70	141.91	153(2)	HMWR	138(14)	TF
				125(10)	TF		
				145	BKM		
5 <sup>3</sup> P	225.56	224.85	221.30	180(30)	TF	225(23)	WSG
				166	BKM		

TABLE II. (Continued).

State	This work			Experimental		Other theoretical	
	0 K	300 K	600 K				
$6^3P$	347.68	344.95	333.84	360(20)	TF	350(35)	WSG
$7^3P$	514.32	506.01	480.45	500(150)	TF	526(53)	WSG
$8^3P$	733.16	712.82	664.59				
$9^3P$	1010.07	968.26	888.09				
$10^3P$	1349.11	1273.70	1151.52				
$11^3P$	1777.62	1650.37	1471.00				
$12^3P$	2282.40	2083.90	1834.05				
$13^3P$	2888.33	2592.57	2254.32				
$14^3P$	3591.94	3172.50	2730.89				
$15^3P$	4403.25	3830.55	3268.04				
$16^3P$	5352.78	4589.26	3882.93				
$17^3P$	6444.09	5450.22	4577.07				
$18^3P$	7661.30	6409.04	5352.96				
$19^3P$	9056.06	7514.58	6254.25				
$20^3P$	10629.99	8800.76	7327.44				
$21^3P$	12452.42	10338.71	8640.22				
$22^3P$	12596.92	11177.24	9823.93				
$3^3D$	14.18	14.18	14.18	14.25(34)	BHR	14.16	WSG
				19.4(5)	TF		
				13.4(5)	IR		
				14(3)	CBHG		
				13.3(1.5)	MBBBLBB		
				17(3)	D		
				13(2)	OV		
				15(2)	PH		
				13(3)	DGJ		
				25(5)	FHJC		
				10(5)	HWR		
$4^3D$	32.07	32.07	32.06	32.1(1.3)	BHR	31.5(1.0)	WSG
				28(10)	TF		
				35.5(1.2)	B		
				32(1)	CBHG		
				27(3)	MBBBLBB		
				42(4)	D		
				23(2)	OV		
				37(6)	PH		
				29(3)	DGJ		
$5^3D$	60.89	60.88	60.68	57.2(2.3)	BHR	60.9(1.9)	WSG
				132(?)	TF		
				55(5)	MBBBLBB		
				59(8)	D		
				35(3)	OV		
				42(5)	DGJ		
$6^3D$	103.29	103.17	102.20	156(?)	TF	107(11)	WSG
$7^3D$	161.92	161.33	158.60	160	TF	145(15)	WSG
$8^3D$	239.44	237.63	231.86	200(50)	TF	207(21)	WSG
$9^3D$	338.59	334.37	323.97	300(100)	TF	300(30)	WSG
$10^3D$	461.97	453.67	436.73	340(60)	TF	380(40)	WSG
$11^3D$	612.25	597.69	571.95				
$12^3D$	792.15	768.59	731.31				
$13^3D$	1004.26	968.37	916.45				
$14^3D$	1251.42	1199.66	1130.18				
$15^3D$	1535.64	1463.84	1373.32				
$16^3D$	1860.33	1764.22	1649.07				
$17^3D$	2228.47	2103.82	1960.51				
$18^3D$	2641.49	2484.58	2310.04				
$19^3D$	3100.51	2909.59	2702.29				



TABLE II. (Continued).

State	This work			Experimental	Other theoretical	
	0 K	300 K	600 K			
$20^3D$	3611.68	3387.83	3148.05			
$21^3D$	4102.41	3872.13	3624.69			
$22^3D$	4641.02	4449.15	4217.63			
$4^3F$	72.27	72.27	72.19	71.6(3.0) BHR		71.9(7.2) WSG
$5^3F$	139.60	139.56	138.21			
$6^3F$	238.52	237.83	231.17			
$7^3F$	375.01	371.30	353.47	400(50) TF		370 TF
$8^3F$	555.16	543.76	508.10			
$9^3F$	784.96	758.93	697.80	750(100) TF		750 TF
$10^3F$	1070.75	1020.77	925.46	1100(200) TF		1100 TF
$11^3F$	1418.16	1332.49	1193.12	1350(300) TF		1270 TF
$12^3F$	1833.08	1697.43	1502.82			
$13^3F$	2321.58	2119.05	1856.74			
$14^3F$	2892.76	2604.89	2263.00			
$15^3F$	3538.87	3147.65	2715.15			
$16^3F$	4297.07	3777.14	3235.91			
$17^3F$	5143.99	4476.19	3814.34			
$18^3F$	6095.58	5261.36	4465.92			
$19^3F$	7157.92	6145.41	5206.05			
$20^3F$	8336.83	7151.18	6064.48			
$21^3F$	9625.18	8310.14	7091.73			
$22^3F$	10912.34	9711.42	8470.79			

(c)

ACLMM	G. Astner, L. J. Curtis, L. Liljeby, S. Mannervik, and I. Martinson, <i>Z. Phys. A</i> <b>279</b> , 1 (1976).
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AW	M. T. Anderson and F. Weinhold, <i>Phys. Rev. A</i> <b>9</b> , 118 (1974).
AJS	L. Allen, D. G. C. Jones, and D. G. Schofield, <i>J. Opt. Soc. Am.</i> <b>59</b> , 842 (1969).
B	K. Buchhaup, <i>Z. Naturforsch.</i> <b>27a</b> , 572 (1972).
BD	R. G. Bennet and F. W. Dalby, <i>J. Chem. Phys.</i> <b>31</b> , 434 (1959).
BHR	H. H. Bukow, G. Heine, and M. Reinke, <i>J. Phys. B</i> <b>10</b> , 2347 (1977).
BJ	W. Bachmann and W. Janke, <i>Z. Naturforsch.</i> <b>28a</b> , 1821 (1973).
BK1	K. A. Bridgett and T. A. King, <i>Proc. Phys. Soc. London</i> <b>92</b> , 75 (1967).
BK2	K. A. Bridgett and T. A. King, in <i>Proceedings of the International Conference on Optical Pumping and Atomic Line Shapes, Warsaw, 1968</i> (unpublished), p. 359.
BKM	W. R. Bennet, P. J. Kindlman, and G. N. Mercer, <i>Appl. Opt. Suppl.</i> <b>2</b> , 34 (1965).
BL	J. M. Berger and A. Lurio, <i>Phys. Rev. A</i> <b>3</b> , 64 (1971).
CBD	C. W. T. Chien, R. E. Bardsley, and F. W. Dalby, <i>Can. J. Phys.</i> <b>50</b> , 116 (1972).
CBHG	S. A. Chin-Bing, C. E. Head, and A. E. Green, <i>Am. J. Phys.</i> <b>38</b> , 352 (1970).
CDDG	M. Carre, J. Desesquelles, M. Dufay, and M. L. Gaillard, <i>Phys. Rev. Lett.</i> <b>27</b> , 1407 (1971).
CMC	M. Cohen and R. P. McEachran, <i>Can. J. Phys.</i> <b>50</b> , 1363 (1972).
D	J. P. Descoubes, <i>Physics of One- and Two-Electron Atoms</i> , edited by F. Bopp and H. Kleinpoppen (North-Holland, Amsterdam, 1969), p. 341.
DGJ	B. Descomps and C. Galerou-Julienne, <i>C. R. Acad. Sci.</i> <b>261</b> , 916 (1965).
DPPB	B. Descomps, J. C. Pebay-Peyroula, and J. Brossel, <i>C. R. Acad. Sci.</i> <b>251</b> , 941 (1960).
DS	K. R. Devine and A. L. Stewart, <i>J. Phys. B</i> <b>5</b> , 2132 (1972).
DW	W. Drtil, <i>Z. Naturforsch.</i> <b>24a</b> , 350 (1969).
FHJC	R. G. Fowler, T. M. Holzberlein, C. H. Jacobson, and S. J. B. Corrigan, <i>Proc. Phys. Soc. London, Sect. A</i> <b>84</b> , 539 (1964).

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FNPP	A. Faure, O. Nedelec, and J. C. Pebay-Peyroula, <i>C. R. Acad. Sci.</i> <b>256</b> , 5088 (1963).
GH	A. H. Gabriel and D. W. O. Heddle, <i>Proc. R. Soc. London, Ser. A</i> <b>258</b> , 124 (1960).
GJK	L. C. Green, N. C. Johnson, and E. K. Kolchin, <i>Astrophys. J.</i> <b>144</b> , 369 (1966).
HDKKD	A. Hitashi, C. Davies, T. King, S. Kubota, and T. Doke, <i>Phys. Rev. A</i> <b>22</b> , 856 (1980).
HWR	S. Heron, R. W. McWhirter, and E. H. Roderick, <i>Proc. R. Soc. London, Ser. A</i> <b>234</b> , 565 (1956).
IR	R. E. Imhof and F. H. Read, <i>J. Phys. B</i> <b>4</b> , 450 (1971).
JF	A. W. Johnson and R. G. Fowler, <i>J. Chem. Phys.</i> <b>53</b> , 65 (1970).
KB	P. J. Kindlman and W. R. Bennet, <i>Bull. Am. Phys. Soc.</i> <b>8</b> , 87 (1963).
KKC	S. A. Kazantsev, A. Kisling, and M. P. Chaika, <i>Opt. Spektrosk.</i> <b>34</b> , 1227 (1973) [ <i>Opt. Spectrosc. (USSR)</i> <b>34</b> , 714 (1973)].
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KOSB	G. S. Kvater, O. V. Oginets, V. B. Smirnov, and S. A. Bagaev, <i>Opt. Spektrosk.</i> <b>35</b> , 389 (1973) [ <i>Opt. Spectrosc. (USSR)</i> <b>35</b> , 226 (1973)].
KS	G. A. Khayrallah and S. J. Smith, <i>Phys. Rev. A</i> <b>18</b> , 559 (1978).
LMP	M. Larsson, B. Mannfors, and W. R. Pendleton, Jr., <i>Phys. Rev. A</i> <b>28</b> , 3371 (1983).
MB	I. Martinson and W. S. Bickel, <i>Phys. Lett.</i> <b>30A</b> , 524 (1969).
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OV	A. L. Osherovich and Ya. Verolainen, <i>Opt. Spektrosk.</i> <b>24</b> , 162 (1981) [ <i>Opt. Spectrosc. (USSR)</i> <b>24</b> , 81 (1968)].
PH	W. R. Pendleton and R. H. Hughes, <i>Phys. Rev.</i> <b>138</b> , A683 (1965).
SPA	B. Schiff, C. L. Pekeris, and Y. Accad, <i>Phys. Rev. A</i> <b>4</b> , 885 (1971), and references therein.
TF	R. T. Thompson and R. G. Fowler, <i>J. Quant. Spectrosc. Radiat. Transfer</i> <b>15</b> , 1017 (1975).
WSG	W. L. Wiese, M. W. Smith, and B. M. Glennon, <i>Atomic Transition Probabilities</i> , Natl. Bur. Stand. (U.S.), Natl. Stand. Ref. Data Ser. No. 4 (U.S. GPO, Washington, D.C. 1966), Vol. 1.
YHM	J. Yellin, T. Hadeishi, and M. C. Michel, <i>Phys. Rev. Lett.</i> <b>30</b> , 417 (1973).

$$\frac{1}{\tau_m^*} = A_m^* = \sum_{m' < m} A(m \rightarrow m') (\langle N \rangle + 1) + \sum_{m'' > m} \frac{g_{m''}}{g_m} A(m'' \rightarrow m) \langle N \rangle, \quad (6)$$

where  $\langle N \rangle = \langle N(h\nu) \rangle$  is the average number of photons with energy  $h\nu$  in thermal equilibrium at temperature  $T$ :

$$\langle N(h\nu) \rangle = \frac{1}{e^{h\nu/k_B T} - 1} = \frac{1}{e^{|\Delta E|/k_B T} - 1}, \quad (7)$$

$g_m$  is the degeneracy of the  $m$ th state, and  $\tau_m^*$  its effective lifetime. We therefore provide in Table II the lifetime values calculated at room temperature (300 K) and at 600 K. The changes from 0 K can be significant, as seen in this table.

Comparison with experimental results in this case is *not*

straightforward, mainly because the various experimental works do not agree with one another, sometimes not even within their claimed error limits. A thorough graphical comparison between them was made by Bukow *et al.*<sup>16</sup> to which work we refer the reader for more details. Much of the uncertainty in the experimental predictions stems from the inaccurate knowledge of cascade contributions. In this context, our work<sup>23</sup> should be particularly useful to experimentalists. The overall agreement of our values with experimental data is good. The most accurately measured lifetime in He is claimed by Astner *et al.*<sup>26</sup> for  $3^1P$ ,  $\tau = 1.7225 \pm 0.0046$  ns. Our prediction,  $\tau = 1.7272$  ns, compares favorably with this value. This agreement is, in our opinion, not fortuitous, considering the agreement of our values for the other states.

Assuming that an excited hydrogenic state  $nl$  decays exclusively through the transition to the lowest possible

state, its lifetime can be shown<sup>27</sup> to be proportional to  $n^3$ . Even when transitions to all states are considered in hydrogen, this proportionality still holds to a very good approximation. Extending the argument to nonhydrogenic systems, and based on the quantum-defect theory, it is often expected in the literature that the lifetime of a Rydberg state with large principal quantum number  $n$  be given by

$$\tau = \tau_0 (n^*)^\alpha, \quad (8)$$

where  $n^*$  is the state's effective quantum number and  $\alpha$  is a number close to 3. Since it is useful to establish scaling laws, this formula is often employed to fit the experimental data. One should be cautioned, however, against the indiscriminant application of such scaling laws, especially of the expectation that  $\alpha \approx 3$ . The reason is that the decay of a state is *not* always predominantly via transition to the ground, or the lowest available, state. Branching to other states can be sizable, and in the case of strong configuration interaction, dominant. With this caution in mind we plot the lifetime values versus  $n^*$  on a log-log scale in Figs. 1 and 2, for the singlet and triplet states, respectively. There are several comments to be made about these figures.

(a) The lifetimes at 0 K are seen to scale, at large  $n$ , like a power of  $n^*$ . We therefore felt comfortable about performing a least-squares fit to the data between  $n = 15$  and 20. The results are presented in Table III. With the *assumption* that this behavior holds true also at considerably larger  $n$  values, the parameters of this table can be used to predict the corresponding lifetimes.

(b) The blackbody radiation (BBR) effects are of different magnitude for different Rydberg series. As was also shown by Farley and Wing,<sup>25</sup> the BBR decay rates do not follow a simple dependence on  $n^*$ . Therefore, the lifetime versus  $n^*$  curves at temperatures other than absolute zero are not linear overall and an attempt of a fitting

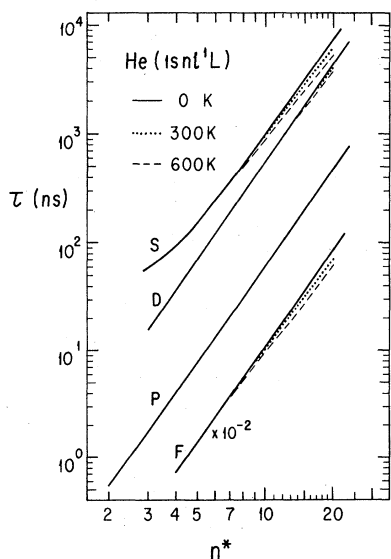


FIG. 1. Lifetimes of the singly excited singlet states  $1s n l {}^1L$  in He.

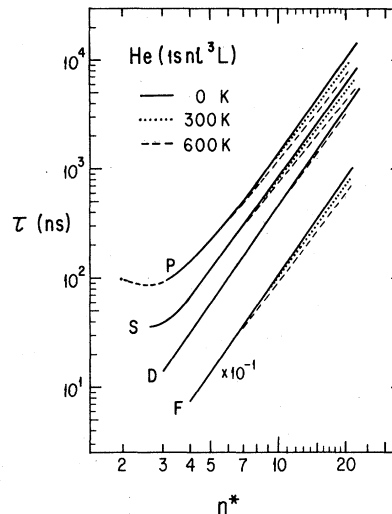


FIG. 2. Lifetimes of the singly excited triplet states  $1s n l {}^3L$  in He.

like Eq. (8) would produce only rough results. At room temperature (300 K) our parameters in Table III can still be used, when augmented by the BBR data of Ref. 25. Above  $n = 20$  the latter data vary smoothly and a simple interpolation is possible at any desired  $n$ . The important point from Table III and from Figs. 1 and 2 is that the exponent  $\alpha$  varies with  $l$  in a nonuniform fashion around 3, is not well defined at nonzero temperatures, and a fit of the experimental data at such temperatures will *always* yield a smaller value than that at 0 K.

(c) The lifetimes of the states with the same  $n$  but different  $l$  quantum numbers are seen to be within a factor of 2 from one another, and their ordering in magnitude not follow the value of  $l$ . An exception to the former point is the behavior of the  ${}^1P$  states, which are seen to be significantly lower than the other states. The obvious reason for this behavior is that  $1s n p {}^1p$  are the only states that can decay directly to the He ground state  $1s^2 {}^1S$ . These transitions of the "principal series" have the largest oscillator-strength values and thus yield the shortest lifetimes.

Based on the overall smooth behavior of the curves in Figs. 1 and 2, the absence below the first ionization potential of any doubly excited states that could drastically alter the character of other states, and the general trend of the various experimental studies, we feel confident in stat-

TABLE III. Values of the parameters  $\alpha$  and  $\tau_0$  appearing in Eq. (8).

	${}^1S$	${}^1P$	${}^1D$	${}^1F$
$\alpha$	2.9224	2.9924	2.9764	2.9690
$\tau_0$ (ns)	1.1742	0.060 548	0.586 33	0.058 881
	${}^3S$	${}^3P$	${}^3D$	${}^3F$
$\alpha$	2.9460	3.0466	2.9764	2.9766
$\tau_0$ (ns)	0.944 74	1.1633	0.485 32	1.1182

ing that from the more recent and systematic investigations, the following experimental determinations of lifetimes are incorrect:

(i) the values of Bukow *et al.*<sup>16</sup> for 3-5<sup>1</sup>S; (ii) the values of Thompson and Fowler<sup>17</sup> for 5-6<sup>1</sup>S, 3,5,6<sup>1</sup>D, 3<sup>3</sup>S, 3<sup>3</sup>P, and 5,6,10<sup>3</sup>D; and, (iii) all values quoted by Hitashi *et al.*<sup>18</sup>

It is our hope that the present work will serve as a use-

ful future guide and will stimulate further experimental investigations.

#### ACKNOWLEDGMENT

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