

Oscillator strengths and radiative branching ratios in atomic Sr

H. G. C. Werij,* Chris H. Greene, C. E. Theodosiou,[†] and Alan Gallagher[‡]
Joint Institute for Laboratory Astrophysics, University of Colorado, Boulder, Colorado 80309;
National Institute of Standards and Technology, Boulder, Colorado 80309;
and Department of Physics, University of Colorado, Boulder, Colorado 80309-0440

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Tables of radiative transition rates (A) are provided for allowed singlet and triplet transitions between Sr states below $n \cong 11$. These are obtained from a combination of R -matrix, multichannel-quantum-defect-theory (MQDT), and modified Coulomb-approximation calculations, plus branching-ratio measurements. Measurements of some spin-changing branchings and of the 4^1D_2 - 5^1S_0 quadrupole transition rate are also reported. Lifetime and A -value measurements from the literature are included in the tables and compared to the calculations. These and the present measurements provide the most comprehensive test to date of calculated transition probabilities for divalent atoms. The MQDT calculation achieves 0–20% agreement with experiment in the great majority of cases tested.

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I. INTRODUCTION

The alkaline-earth-metal atoms have many useful features, such as visible resonance lines and zero-spin isotopes, and they are frequently used in experiments. Interpreting these experiments often requires a knowledge of transition oscillator strengths, which are often poorly known. Several laboratories, including our own, have recently carried out experiments in Sr vapor, whose interpretation requires oscillator strengths for many transitions between excited states. Consequently, we surveyed the literature that includes experiments as well as multiconfigurational Hartree-Fock (MCHF) and Coulomb-approximation (CA) calculations [1]. We hoped to find reasonable consistency between experiments and between experiment and calculated values, but we were frequently disappointed in both comparisons. In the case of the CA, the reasons for frequent problems with alkaline-earth-metal atoms are clear. In spite of the fact that we are considering transitions between excited states, many of these states are not well described as a single configuration, or as a relatively isolated outer electron around a $(ns)^2S$ ion core. Also for this reason the lowest 1D and 3D states of Sr have effective quantum numbers n^* of about 2, precluding a normal CA result for transitions to these levels [1]. Both irregularities in level energies and in oscillator strengths can be understood if one treats both outer electrons beyond the independent electron model. This was done in MCHF calculations by Vaecck, Gedefroid, and Hansen [2] for singlet transitions, and it is done here for singlets and triplets using the R -matrix method with multichannel-quantum-defect theory (MQDT) to include all major interacting channels [3–11]. For additional comparison we have also carried out a modified CA, using a Hartree-Slater core approximation (HSCA).

Previous experimental, and some theoretical, results for Sr oscillator strengths are presented below, where they are compared to the present results. Oscillator strengths for the n^1P to 4^1D_2 transitions were particu-

larly important for the analysis of the data in Ref. [12], so we have carried out an experiment to establish some of these. For several of the n^1P_1 states, we measured the fluorescence branching to the 4^1D_2 state versus the 5^1S_0 (ground) state. The n^1P_1 - 5^1S_0 oscillator strengths f are well known from previous experiments, so these ratios establish the desired n^1P_1 - 4^1D_2 values. Details of this experiment and the results are given below. Measurements of fluorescence branching from some other spin-orbit mixed levels are also reported. Another experiment recently carried out in our laboratory [13] detected ratios of several different adsorptions from the 5^1P , 4^1D , 4^3D , and 5^3P states of Sr. This determined ratios of f for these transitions, but did not directly yield f values. The experiment is not described here, but we include the results below. The f value of the 4^1D_2 to 5^1S_0 quadrupole transition has also been determined from a third type of measurement, which is described below.

In the following sections we first describe the experiments and their results, then the MQDT calculations. The CA calculations followed standard procedures and are not described in detail. Tables of oscillator strengths are given, in which the CA, MQDT, and various experimental results are compared. A brief discussion follows, but it is not feasible to draw very general conclusions. Agreement between theory and experiment, or lack thereof, is evident from reading the tables.

II. MEASUREMENTS

A. Branching ratios

Radiative branching ratios have been determined for transitions from Sr 1P levels and from some strongly configuration-mixed states, by measuring intensity ratios in the emission spectrum of a Sr hollow-cathode lamp. The lamp light was focused on the entrance slit of a $\frac{3}{4}$ -m double monochromator using a quartz lens and detected with a photomultiplier. For any pair of transitions sharing a common upper level i , the ratio of transition proba-

bilities can be found from the relation:

$$\frac{A_{ij}}{A_{ik}} = \frac{S(\lambda_{ij})\epsilon(\lambda_{ik})T(\lambda_{ik})}{S(\lambda_{ik})\epsilon(\lambda_{ij})T(\lambda_{ij})}$$

Here $S(\lambda)$ is the photomultiplier signal for the fluorescence at wavelength λ and $T(\lambda)$ is the transmission of the synthetic silica window of the hollow-cathode lamp. The spectral sensitivity of the detection system per photon $\epsilon(\lambda)$ is determined by geometry, wavelength-dependent transmission of lens and monochromator, and spectral response of the photomultiplier tube.

The first problem that has to be solved is identifying the proper Sr lines. In many wavelength regions the lamp spectrum is extremely complicated, and many of the observed lines have not been identified yet. Since our Sr hollow-cathode lamp contains Ne as a buffer gas, it is often even ambiguous whether a particular line is due to a Sr or a Ne transition. The task of identifying the spectral lines was in some cases complicated by the fact that the reading of the monochromator typically varied up to a 1 Å from one scan to another. In those cases the proper Sr lines were identified by comparing the measured spectrum from the Sr lamp with the known spectrum of a Ne, Kr, or Hg lamp, which was measured simultaneously.

The spectral sensitivity $\epsilon(\lambda)$ is obtained from a calibration using a calibrated tungsten iodide lamp:

$$\epsilon(\lambda) = [CS^W(\lambda)D(\lambda)] / [\lambda I^W(\lambda)]$$

Here $S^W(\lambda)$ is the photomultiplier signal and C is a wavelength-independent factor, which is arbitrary, since only a ratio of spectral sensitivities is used in our analysis. Since the tungsten lamp has a continuous spectrum and atomic lines from the hollow-cathode lamp are monochromatic, the irradiance $I^W(\lambda)$ of the tungsten lamp (in $\text{W}/\text{cm}^2 \text{Å}$) is divided by the monochromator dispersion $D(\lambda)$.

We have determined radiative branching ratios for the α 1P_1 levels connecting to the $5s^2$ 1S_0 ground state and the $5s4d$ 1D_2 metastable state for $\alpha = 5s6p, 5s7p, 4d5p, 5s8p,$ and $5s9p$. A significant difficulty resulted from the fact that the wavelengths of the measured pair of transitions lie in widely separated parts of the spectrum (see Table I). The spectral sensitivity $\epsilon(\lambda)$ varies in some cases more than two orders of magnitude and accurate knowledge of $\epsilon(\lambda)/\epsilon(\lambda')$ is crucial. The largest uncertainties in this ratio occurred for wavelengths below 2500 Å, as we had to extrapolate the available irradiance data of the calibrated tungsten lamp, and the irradiance and spectral sensitivity both drop rapidly in the ultraviolet, causing a poor signal-to-noise ratio. By multiplying the irradiance of a black-body source by the emissivity of tungsten [17], we obtained an excellent fit to the calibration data of the tungsten lamp from 2500–7000 Å and estimate the extrapolation error to be less than 10% at 2300 Å. A maximum uncertainty of ~15% resulted for the weakest signal S^W at 2307 Å.

Another complication was that the throughput of the monochromator was strongly dependent on which parts of the gratings were illuminated. Since the index of refraction of the quartz lens changes considerably with wavelength, proper focusing of the lamp light on the entrance slit of the monochromator did not occur over the entire wavelength interval under study. This might result in a partial filling of the monochromator gratings, which would make the throughput of the monochromator very sensitive to the position of the lamp. Extreme care was taken to ensure that the optical geometry was the same, when calibrating the spectral sensitivity using the tungsten lamp and when determining the ratio $S(\lambda_{ij})/S(\lambda_{ik})$ using the Sr hollow-cathode lamp. The optical path in the monochromator was further defined by installing an aperture in front of the first grating, and the quartz lens was positioned to optimally focus the lamp onto the slit for $\lambda \sim 3100$ Å, at which wavelength

TABLE I. Transition probabilities for n $^1P \rightarrow 5s^2$ $^1S, 4s4d$ 1D . (1): Upper level, decaying to $5s^2$ 1S and $5s4d$ 1D ; (2) detected wavelengths; (3) experimental A coefficient for $^1P \rightarrow 5s^2$ 1S (Parkinson, Reeves, and Tomkins [14]); (4) present R -matrix calculation, (5) experimental ratio of A coefficients, this work; (6) column (3) \times column (5), except values marked with superscript a or b . Here and in the subsequent tables, underlined values are obtained from a combined MQDT and variational R -matrix calculation.

Upper level	λ_{air} (Å)	$A_{\text{expt}}(\rightarrow 5^1S)$ (10^7 s^{-1})	$A_{\text{theor}}(\rightarrow 5^1S)$ (10^7 s^{-1})	$\frac{A(\rightarrow 4^1D)}{A(\rightarrow 5^1S)}$	$A_{\text{expt}}(\rightarrow 4^1D)$ (10^7 s^{-1})	$A_{\text{theor}}(\rightarrow 4^1D)$ (10^7 s^{-1})
(1)	(2)	(3)	(4)	(5)	(6)	(4)
$5s5p$ 1P	4607.3 64 600	20.1±0.6	<u>2.15</u>		3.9×10^{-4a}	<u>1.7×10^{-3}</u>
$5s6p$ 1P	2931.8 7167.2	0.186±0.03 0.18 ^b	<u>0.379</u>	5.0±0.7	0.94±0.20 0.57 ^b	<u>1.56</u>
$5s7p$ 1P	2569.5 5329.8	0.54±0.07 0.37 ^b	<u>0.319</u>	3.1±0.4	1.7±0.3 2.36 ^b	<u>2.16</u>
$4d5p$ 1P	2428.1 4755.5	1.69±0.23 0.67 ^b	<u>1.20</u>	1.25±0.18	2.1±0.4	<u>2.29</u>
$5s8p$ 1P	2354.3 4480.5	1.84±0.24	<u>1.49</u>	1.02±0.16	1.9±0.4	<u>1.78</u>
$5s9p$ 1P	2307.4 4313.2	1.17±0.17	<u>1.16</u>	1.1±0.3	1.3±0.4	<u>1.06</u>

^aHunter, Walker, and Weiss (Ref. [15]) ±40% uncertainty.

^bCorliss and Bozman, Ref. [16] ±40% uncertainty estimated here.

the lens index of refraction and focal length have an average value for the wavelength range under study.

In order to check the reliability of our procedure we measured the $5s7p\ ^1P$ branching ratio both with and without the quartz lens. The results agreed within 3%, well within the overall uncertainty given in column (5) of Table I. The intensities of the ultraviolet lines of the higher-lying 1P levels were so weak that they only could be measured with the quartz lens.

When measuring the relative intensity of spectral lines, it is important to ascertain if radiation trapping in the lamp has altered the observed intensity ratios. In particular, trapping of the $n\ ^1P \rightarrow 5s^2\ ^1S$ fluorescence could play a significant role. In order to check for such effects the intensity ratios of $n\ ^1P \rightarrow 5s4d\ ^1D$ to $n\ ^1P \rightarrow 5s^2\ ^1S$ fluorescence have been measured for various values of the discharge current through the lamp, ranging from 5.5 mA to 18.4 mA. For the $5s6p$, $5s7p$, $4d5p$, and $5s8p\ ^1P_1$ levels a dependence of the intensity ratio on the current was observed, and the branching ratios in the absence of radiation trapping were obtained from an extrapolation to zero lamp current. The extrapolated ($5\ ^1S_0; 4\ ^1D_2$) branching ratios at 0 mA were 16–21% smaller than that measured at 18.4 mA, indicating significant radiation trapping of the $n\ ^1P \rightarrow 5s^2\ ^1S$ fluorescence. For the

$5s9p\ ^1P$ level no clear dependence on the current could be determined, since the signal-to-noise ratio rapidly decreased with decreasing current.

Column (5) of Table I contains the measured ratios of radiative decay rates for the transitions $n\ ^1P \rightarrow 5s4d\ ^1D$ and $n\ ^1P \rightarrow 5s^2\ ^1S$, as obtained from the extrapolation to zero lamp current. From the known $n\ ^1P \rightarrow 5s^2\ ^1S$ transition probabilities [column (3), Ref. [14]] we calculate the $n\ ^1P \rightarrow 5s4d\ ^1D$ decay rates given in column (6) of Table I. Transition probabilities for the $5s5p\ ^1P$ upper level (not measured here) have been included in the table.

In order to determine the singlet or triplet character of the strongly spin-orbit-mixed states $5s6p\ ^1P_1$, $5s6p\ ^3P_J$, $4d5p\ ^1D_2^0$, and $4d5p\ ^3F_J$ radiative branching ratios from these states to the $5s4d\ ^1D_2$ and $5s4d\ ^3D_J$ states have also been measured. The procedure is the same as outlined above, but most of the difficulties discussed above do not occur since the observed wavelengths in this case lie relatively close together in the visible part of the spectrum. Relative transition probabilities (in arbitrary units) are given in Table II. It should be noted that all transitions from a given upper level are not necessarily listed here and that only values for transitions having a common upper level should be compared.

TABLE II. Measured transition probability ratios involving spin-forbidden transitions.

Upper level	Lower Level	$\lambda_{\text{air}}\ (\text{\AA})$	A (relative units)
$5s6p\ ^1P_1$	$5s4d\ ^1D_2$	7167.2	92.8 ± 1.8^a
	$5s4d\ ^3D_1$	6270.4	0.14 ± 0.03
	$5s4d\ ^3D_2$	6294.0	$< 7.0^b$
$5s6p\ ^3P_0$	$5s4d\ ^3D_1$	6369.9	100 ^c
$5s6p\ ^3P_1$	$5s4d\ ^3D_1$	6363.9	19.5 ± 0.4
	$5s4d\ ^3D_2$	6388.2	71.9 ± 1.5
	$5s4d\ ^1D_2$	7287.4	8.5 ± 0.2
$5s6p\ ^3P_2$	$5s4d\ ^3D_1$	6321.8	0.51 ± 0.02
	$5s4d\ ^3D_2$	6345.7	11.0 ± 0.2
	$5s4d\ ^3D_3$	6386.5	56.6 ± 1.1
	$5s4d\ ^1D_2$	7232.1	31.9 ± 0.7
$4d5p\ ^1D_2^0$	$5s4d\ ^1D_2$	7309.4	85.3 ± 1.7^d
	$5s4d\ ^3D_1$	6380.7	11.4 ± 0.2
	$5s4d\ ^3D_2$	6405.1	0.06 ± 0.01
	$5s4d\ ^3D_3$	6446.7	3.2 ± 0.1
$4d5p\ ^3F_2$	$5s4d\ ^3D_1$	6617.3	64.9 ± 1.3^e
	$5s4d\ ^3D_2$	6643.5	18.4 ± 0.6
	$5s4d\ ^3D_3$	6688.2	0.24 ± 0.17
	$5s4d\ ^1D_2$	7621.5	< 17
$4d5p\ ^3F_3$	$5s4d\ ^3D_2$	6504.0	84 ± 3
	$5s4d\ ^3D_3$	6546.8	15.9 ± 0.6
	$5s4d\ ^1D_2$	7438.4	?(overlap)
$4d5p\ ^3F_4$	$5s4d\ ^3D_3$	6408.5	100

^aIn Table I, 93 in these units corresponds to $A = 9.4 \times 10^6/\text{s}$.

^bAn upper limit is given when overlapping lines were unresolved.

^cUsing 3P_J averaged branching in Table VIII, ~ 80 in these units corresponds to $A = 15 \times 10^6/\text{s}$.

^dIn Table XI, 85 in these units corresponds to $A = 39 \times 10^6/\text{s}$.

^eIn Table X, 100 in these units corresponds to $A = 24 \times 10^6/\text{s}$.

B. Quadrupole transition

The $5s4d^1D_2 \rightarrow 5s^2S_0$ quadrupole transition was studied in a different experimental setup using a double-window Sr cell, which is described in Ref. [12]. A Nd:YAG (YAG denotes yttrium-aluminum-garnet) pumped dye laser (pulses 6 ns, 0.1 mJ) excited the $5s5p^1P_1$ state, which had an effective lifetime of $\sim 0.6 \mu\text{s}$ due to strong radiation trapping. When the Sr($5s5p^1P$) density is produced above a certain threshold, which depends on Sr density, laser power, and detuning, the metastable $5s4d^1D$ state becomes strongly populated during the laser pulse by stimulated $5s5p^1P \rightarrow 5s4d^1D$ emission. In our experiment the effective lifetime of the $5s4d^1D$ state was $\sim 12 \mu\text{s}$, due to wall collisions.

The densities of the $5s5p^1P$ and $5s4d^1D$ levels were obtained from the absorption of spectral lines from a Sr hollow-cathode lamp, connecting those levels to the levels $5s5d^1D_2$ and $4d5p^1D_2^0$, respectively [12]. The same $\frac{3}{4}$ -m double monochromator and photomultiplier were used to detect the lamp light and the fluorescence emitted by the Sr vapor in the cell. The amplified photomultiplier output was processed by a transient digitizer and the time-resolved signals were typically averaged over 10^3 – 10^4 pulses.

Under conditions such that stimulated production of the $5s4d^1D$ state occurred, strong fluorescence around 4962 Å was observed. Several Sr transitions occur near this wavelength and might contribute to the observed signal: $5s4d^1D_2 \rightarrow 5s^2S_0$ (4961.5 Å), $5s5d^3D_3 \rightarrow 5s5p^3P_2$ (4962.1 Å) and $5s7d^1D_2 \rightarrow 5s5p^1P_1$ (5965.5 Å). The monochromator resolution of ~ 5 Å was insufficient to separate these at the 1-mm slit widths used to obtain

good signal-to-noise ratio. However, the following observation showed that this signal was dominated by the 4^1D_2 – 5^1S_0 quadrupole radiation. The $5s5d^3D_3$ and $5s7d^1D_2$ levels were populated as a result of energy-pooling collisions between Sr atoms in the $5s5p^1P$ and $5s4d^1D$ states and/or subsequent cascading from more highly excited states that are similarly populated [12]. As the $5s5p^1P$ state decays at a ~ 0.5 - μs rate, only collisions between pairs of $5s4d^1D$ states remain after a few μs . If any of the observed fluorescence (after a few μs) were due to direct or cascade populating the $5s5d^3D_3$ or $5s7d^1D_2$ levels by energy-pooling collisions, it would decay at least twice as fast as the $5s4d^1D$ population. However, the observed ~ 4962 -Å fluorescence closely followed the 12- μs decay of the $5s4d^1D$ density, hence it is mainly due to the $5s4d^1D_2 \rightarrow 5s^2S_0$ quadrupole transition. In a second test we varied the laser detuning and intensity to alter the $5s4d^1D$ density from $2 \times 10^{10} \text{ cm}^{-3}$ to $6 \times 10^{11} \text{ cm}^{-3}$. The 4962-Å fluorescence, measured at 2 μs and 16 μs after the laser pulse, was a linear function of the $5s4d^1D$ density, further confirming the source of this radiation. The 4962-Å fluorescence signal S_q is then given by

$$S_q = A_{D-S} n_D \epsilon(\lambda_q),$$

where A_{D-S} is the radiative decay rate of the quadrupole transition, n_D is the $5s4d^1D_2$ density, and $\epsilon(\lambda)$ is the detection sensitivity. Likewise we can write for the $5s5p^1P_1$ state resonance fluorescence:

$$S_r = \Gamma_{\text{eff}} n_P \epsilon(\lambda_r),$$

where Γ_{eff} is the effective decay rate of the $5s5p^1P_1$ den-

TABLE III. Theoretical transition probabilities (10^6 s^{-1}). Underlined values are based upon a combined MQDT and variational R -matrix calculation, the others are calculated using the HSCA approximation. Experimental A values are in parenthesis, and theoretical and experimental lifetimes are given at the bottom. Several labels are different from those given in Moore's tables (Ref. [23]). In the following tables all levels are listed in order of increasing energy, which should eliminate any possible ambiguity.

Lower level	Upper level							
	$5s6s^1S$	$5p^2^1S$	$5s7s^1S$	$5s8s^1S$	$5s9s^1S$	$5s10s^1S$	$5s11s^1S$	
$5s5p^1P$	<u>18.6</u>	<u>29.3</u> (36) ^a	<u>47.2</u> (38) ^a	<u>7.44</u>	<u>3.35</u>	<u>1.88</u>	<u>1.17</u>	
$5s6p^1P$		<u>0.009</u>	5.59	2.67	1.41	0.85	0.60	
$5s7p^1P$				1.92	0.94	0.55	0.41	
$4d5p^1P$					0.0	0.0	0.0	
$5s8p^1P$					0.011	0.051	0.070	
$5s9p^1P$						0.044	0.004	
$5s10p^1P$							0.044	
$\sum A$	<u>18.6</u>	<u>29.3</u>	<u>52.8</u>	<u>12.0</u>	<u>5.71</u>	<u>3.38</u>	<u>2.30</u>	present
τ_{theor} (ns)	<u>53.8</u>	<u>34.1</u>	<u>19</u>	<u>83</u>	<u>175</u>	<u>296</u>	<u>435</u>	present
τ_{expt} (ns)	35.9	31.0	68	112	212	340	470	HSCA ^b
		28	23					a
		67.6±3.5	18.4±1.1	70.9±5.4	119±5	303±26	376±31	c
			20±2	86±4	128±4			d

^aCorliss and Bozman, Ref. [16] $\pm 40\%$ estimated uncertainty.

^bUsing Coulomb approximation for all transitions.

^cGornik, Ref. [24].

^dOsherovich *et al.*, Ref. [25].

sity n_p , determined by radiation trapping. The quadrupole transition probability A_{D-S} can thus be expressed as

$$A_{D-S} = \frac{\epsilon(\lambda_r) S_q \Gamma_{\text{eff}} n_p}{\epsilon(\lambda_q) n_D S_r}.$$

We determined the ratio S_q/n_D from the data with an uncertainty of $\sim 10\%$. Under the same conditions we measured the ratio $S_r(\Gamma_{\text{eff}} n_p)$ for the resonance line. This quantity typically fluctuated by $\pm 12\%$ but did not show any systematic behavior as a function of the varied pa-

rameters. In addition to these experimental uncertainties, systematic errors in the densities n_p and n_D result from uncertainties of $\sim 10\%$ in the transition probabilities used to calculate these densities from absorption measurements.

From the measurements discussed above the radiative decay rate A_{D-S} is $51 \pm 12 \text{ s}^{-1}$, which agrees within experimental uncertainty with the theoretical values of 45 s^{-1} by Bauschlicher, Langhoff, and Partridge [18] and 43 s^{-1} by Vaeck, Gedefroid, and Hansen [2]. The present HSCA calculation obtains $A_{D-S} = 33 \text{ s}^{-1}$. (The value obtained in

TABLE IV. Theoretical transition probabilities (10^6 s^{-1}) and related measurements. Underlining, parentheses, and HSCA as in Table III.

Lower level	Upper level								
	$5s5p \ ^1P$	$5s6p \ ^1P$	$5s7p \ ^1P$	$4d5p \ ^1P$	$5s8p \ ^1P$	$5s9p \ ^1P$	$5s10p \ ^1P$	$5s11p \ ^1P$	
$5s^2 \ ^1S$	<u>215</u> 174 ^a	<u>3.79</u> 1.06 ^a (1.86±0.03) ^b	<u>3.19</u> 5.5 ^a	<u>12.0</u> 14 ^a	<u>14.9</u> 14.6 ^a	<u>11.6</u>	<u>7.60</u>	<u>4.88</u>	
$5s6s \ ^1S$		<u>2.67</u>	<u>1.08</u>	<u>2.93</u>	<u>2.66</u>	<u>1.61</u>	<u>0.86</u>	<u>0.47</u>	
$5p^2 \ ^1S$			<u>0.070</u>	<u>0.010</u>	<u>0.021</u>	<u>0.067</u>	<u>0.077</u>	<u>0.067</u>	
$5s7s \ ^1S$			0.033	0.000	0.74	0.20	0.10		
$5s8s \ ^1S$				0.000	0.49	0.27	0.13		
$5s9s \ ^1S$						0.23	0.10		
$5s10s \ ^1S$							0.11		
$5s4d \ ^1D$	0.017 0.005 ^a	<u>15.6</u> 12.9 ^a (9.4) ^b , (5.7) ^c	<u>21.6</u> 18.3 ^a (23) ^c	<u>22.9</u> 19.5 ^a	<u>17.8</u> 14.5 ^a	<u>10.6</u>	<u>5.79</u>	<u>3.27</u>	
$5s5d \ ^1D$			<u>0.71</u> 1.06 ^a	<u>0.49</u> 0.75 ^a	<u>0.20</u> 0.48 ^a	<u>0.070</u>	<u>0.025</u>	<u>0.010</u>	
$5p^2 \ ^1D$			0.000	0.006 0.20 ^a	0.000 0.06 ^a	0.000	0.000	0.000	
$5s6d \ ^1D$					0.16	0.17	0.000		
$5s7d \ ^1D$					0.25	0.024	0.001		
$5s8d \ ^1D$						0.077	0.003		
$5s9d \ ^1D$							0.031		
$5s4d \ ^3D$		(< 0.7) ^c							
$\sum A$	<u>215</u>	<u>22.1</u>	<u>26.7</u>	<u>38.3</u>	<u>37.2</u>	<u>24.9</u>	<u>14.8</u>	<u>8.70</u>	present
τ_{theor} (ns)	<u>4.65</u>	<u>45.3</u>	<u>37.5</u>	<u>26.1</u>	<u>26.9</u>	<u>40.1</u>	<u>67.6</u>	<u>115</u>	present
τ_{expt} (ns)	4.32	143	188	9.9	45	70	125		HSCA
		68 ^f							b, c, e
		65±5	39.2±2.0	23.4±2.3	27.1±1.3	42.7±2.0	81±6	125±9	g
	4.97±0.15								h
	4.56±0.21								i
	5.29±0.10								j
	5.4±0.5								k
								92±5	l
									m
								110±10	

^aVaeck, Gedefroid, and Hansen, Ref. [2]. MCHF calculation.

^bParkinsen, Reeves, and Tomkins, Ref. [14] and Table I.

^cMiller *et al.*, Ref. [11] $\pm 20\%$ uncertainty.

^dCorliss and Bozman, Ref. [16].

^eTable II.

^fIncluding the calculated $5s6s \ ^1S$ transition.

^gHulpke, Paul, and Paul, Ref. [28].

^hDickie *et al.*, Ref. [29].

ⁱJönsson *et al.*, Ref. [26].

^jLurio, DeZafra, and Goshen, Ref. [27].

^kErdevdi and Shimon, Ref. [30].

^lGrafström *et al.*, Ref. [31].

^mSchwenz and Leone, Ref. [32].

Ref. [18] for the comparable electric quadrupole transition in calcium is also in excellent agreement with the experimental result reported by Fukuda and Ueda [19].)

III. CALCULATIONS

Strontium exhibits strong configurational mixing and significant spin-orbit mixing, affecting energies as well as the oscillator strengths. The CA calculation of oscillator strengths assume simple one-electron transitions between single configurations, usually $5snl$, and is most appropriate for transitions between well excited states. The CA cannot be directly used for the 4^3D and 4^1D states, as they are too strongly bound ($n^* \cong 2$). To handle this problem, we have used a modified CA, based on a Hartree-Slater core. We have applied this to all transitions of interest, ignoring lesser components of the states with mixed configurations. To include configuration-mixing effects, these transition probabilities have also been calculated by MQDT and R matrix, for transitions to the seven lowest states (5^1S , 5^3P , 4^3D , 4^1D , 5^1P , 6^1S , 6^3S). (For brevity we refer to the combined R -matrix and MQDT procedure as MQDT throughout this paper.) These values are reported in Tables I–X, where they are

underlined to distinguish them from HSCA results in the same tables.

A. Coulomb approximation with a Hartree-Slater core (HSCA)

The philosophy of the Coulomb approximation [1] is the fact that the Rydberg state wave functions, at least for medium to high principal quantum numbers n , are determined by their energies, or equivalently, their effective quantum numbers n^* . Apart from its effect on n^* , i.e., the introduction of a “quantum defect,” the ion core affects the wave functions only at small distances, and for a dipole transition this is usually a small effect. Therefore the wave functions can be approximated either by analytic Whittaker wave functions with the appropriate n^* , or by inward numerical integration and a judicious cutoff of the integration at small distances. This approach has been augmented by one of us [20] to obtain the radial wave function by inward integration of the single-electron radial Schrödinger equation, using $V(r)$ from a Hartree-Slater self-consistent field calculation using standard programs [21] and energy values from experiment or, where necessary, from extrapolation or interpolation to levels not observed experimentally. This approach was developed to treat alkalilike and heliumlike systems where the Rydberg series are not strongly perturbed [20,22].

TABLE V. Theoretical transition probabilities (10^6 s^{-1}) and related measurements. Underlining, parentheses, and HSCA as in Table III.

Lower level	Upper level						
	$5s5d^1D$	$5p^2^1D$	$5s6d^1D$	$5s7d^1D$	$5s8d^1D$	$5s9d^1D$	
$5s5p^1P$	<u>9.28</u> (6.8) ^a	<u>64.7</u> (46) ^a	<u>21.8</u> (12.6) ^a	<u>6.64</u>	<u>2.14</u>	<u>0.555</u>	
$5s6p^1P$	<u>0.009</u>	0.000	3.52	2.51	1.64	1.10	
$5s7p^1P$			0.24	0.44	0.47	0.40	
$4d5p^1P$				0.000	0.000	0.000	
$5s8p^1P$					0.25	0.023	
$5s9p^1P$						0.16	
$4d5p^1D^0$		0.017	0.000	0.000	0.000	0.000	
$4d5p^1F$			0.000	0.000	0.000	0.000	
$5s4f^1F$			0.003	0.035	0.024	0.021	
$5s5f^1F$				0.027	0.001	0.003	
$5s6f^1F$					0.013	0.002	
$5s7f^1F$						0.004	
$\sum A$	<u>9.29</u>	<u>64.7</u>	<u>25.6</u>	<u>9.65</u>	<u>4.54</u>	<u>2.27</u>	present
τ_{theor} (ns)	<u>108</u> 20.4	<u>15.5</u> 32.4	<u>39</u> 49.3	<u>104</u> 90	<u>220</u> 149	<u>441</u> 243	present HSCA
τ_{expt} (ns)	115±5.8	23.8±1.3 25±3	42.6±2.2 44±3	114.5±4 77±5	182±16 204±10	413±46	b c d
	147	22	61				a and HSCA

^aCorliss and Bozman, Ref. [16], ±40% estimated uncertainty.

^bGornik, Ref. [24].

^cOsherovich *et al.* Ref. [25].

^dErdevdi and Shimon, Ref. [30].

B. MQDT, R-matrix calculations

Rapid strides in recent years have dramatically improved our ability to describe valence spectra of the heavy alkaline-earth-metal atoms [3–6], namely Ca, Sr, Ba, and Ra. A major problem in treating these atoms heavier than argon is the extreme sensitivity of the effective potential barrier for *d* and *f* electrons to the independent-electron approximation used to describe inner-shell-electron screening effects. This sensitivity causes large errors in *ab initio* calculations, even of one-electron energy levels, e.g., of Sr⁺. These errors in the one-electron levels propagate in turn and frequently cause calculated two-electron spectra to bear little resemblance to experiment.

The main element needed to alleviate this difficulty was recognized long ago by Laughlin and Victor [7]. A one-electron model potential $V(r)$ is introduced, which con-

tains adjustable parameters (see Table I of Ref. [6]) chosen semiempirically to give accurate energy levels, e.g., of Sr⁺. This constraint on the one-electron potential guarantees that the de Broglie phase of an electron emerging from the Sr²⁺ core will be correct. The complicated, highly perturbed two-electron valence spectra of Sr can then be described to near-spectroscopic accuracy by the following “two-electron” model Hamiltonian,

$$H = -\frac{1}{2}\nabla_1^2 - \frac{1}{2}\nabla_2^2 + V(r_1) + V(r_2) + \frac{1}{r_{12}}. \quad (1)$$

Numerous calculations using this Hamiltonian have now shown that the resulting observables (including energy levels, photoionization cross sections, autoionization profiles, photoelectron angular distributions, etc.) are nearly independent of the one-electron potential used, as long as the one-electron energy levels are accurate.

TABLE VI. Theoretical transition probabilities (10^6 s^{-1}) and related measurements. Underlining, parentheses, and HSCA as in Table III.

Lower level	Upper level						
	$4d5p \ ^1F$	$5s4f \ ^1F$	$5s5f \ ^1F$	$5s6f \ ^1F$	$5s7f \ ^1F$	$5s8f \ ^1F$	
$5s4d \ ^1D$	<u>2.08</u> 0.25 ^a	<u>15.7</u> 24 ^a (26) ^b (22) ^c	<u>16.8</u> 18 ^a	<u>11.6</u> 11.2 ^a	<u>7.87</u> 7.2 ^a	<u>5.48</u>	
$5s5d \ ^1D$	0.000 1.33 ^a	11.5 3.4 ^a	2.15 0.59 ^a	0.60 0.08 ^a	0.23 0.009 ^a	0.036	
$5p^2 \ ^1D$	0.003 0.009 ^a	0.000 1.1 ^a	0.000 0.014 ^a	0.000 0.010 ^a	0.000 0.020 ^a	0.000	
$5s6d \ ^1D$			2.10	0.50	0.18	0.039	
$5s7d \ ^1D$				0.73	0.26	0.10	
$5s8d \ ^1D$					0.30	0.13	
$5s9d \ ^1D$						0.16	
$5s5g \ ^1G$				0.18	0.009	0.008	
$5s6g \ ^1G$					0.019	0.015	
$5s7g \ ^1G$						0.024	
$\sum A$	<u>2.08</u>	<u>27.2</u>	<u>21.1</u>	<u>13.4</u>	<u>8.87</u>	<u>5.99</u>	present
τ_{theor} (ns)	<u>480</u> 12.9	<u>36.8</u> 72.9	<u>47.5</u> 98.2	<u>74.4</u> 159	<u>113</u> 242	<u>167</u> 298	present HSCA
τ_{expt} (ns)		33.5±1.2 29.7±0.9	33.6±1.3 53±2	98.5±12 81±8	133±4 120±5 126±5	228±8 179±16	d e f g h i
	296±22	31.3±0.9 34.3±2.8 34.2±0.4 27	45.0±2.1	78±3			a and HSCA

^a Vaecq, Gedefroid, and Hansen, Ref. [2], MCHF calculation.

^b Corliss and Bozman, Ref. [16].

^c Miller *et al.*, Ref. [13].

^d Gornik, Ref. [24].

^e Osherovich *et al.*, Ref. [25].

^f Jönsson *et al.*, Ref. [26].

^g Grafström *et al.*, Ref. [31].

^h Brinkmann, Ref. [33].

ⁱ Andrä *et al.*, Ref. [34].

TABLE VII. Theoretical transition probabilities (10^6 s^{-1}) and related measured lifetimes. Underlining and HSCA as in Table III.

Lower level	Upper level					
	$5s6s \ ^3S$	$5s7s \ ^3S$	$5s8s \ ^3S$	$5s9s \ ^3S$	$5s10s \ ^3S$	
$5s5p \ ^3P$	<u>71.3</u>	17.5 (12) ^a	8.22	4.53	2.77	
$5s6p \ ^3P$		6.97	2.53	1.33	0.79	
$5s7p \ ^3P$			1.61	0.56	0.31	
$5s8p \ ^3P$				0.63	0.24	
$5s9p \ ^3P$					0.28	
$\sum A$	<u>71.3</u>	24.5	12.4	7.05	4.39	present
τ_{theor} (ns)	<u>14.0</u>					MQDT
	19.0	41	81	142	228	HSCA
τ_{expt} (ns)		34.8±1.3				b
		40.0±3.4				c
	15.0±0.8	34.2±1.7				d
	10.9±1.1					e
	12.9±0.7					f
		52				a and HSCA

^aCorliss and Bozeman, Ref. [16].

^bGornik, Ref. [24].

^cOsheroich *et al.*, Ref. [25].

^dJönsson *et al.*, Ref. [26].

^eBrinkmann, Ref. [33].

^fHavey, Balling, and Wright, Ref. [35].

^gUeda, Ashizawa, and Fukuda, Ref. [36].

TABLE VIII. Theoretical transition probabilities (10^6 s^{-1}) and related measurements. Underlining and HSCA as in Table III. The $5 \ ^3P_1$ state lifetime, not shown, has been measured to be $\sim 21 \ \mu\text{s}$ (Refs. [37–39]).

Lower level	Upper level					
	$5s6p \ ^3P$	$4d5p \ ^3P$	$5s7p \ ^3P$	$5s8p \ ^3P$	$5s9p \ ^3P$	
$5s6s \ ^3S$	<u>6.73</u>	<u>0.70</u>	<u>0.84</u>	<u>0.15</u>	<u>0.040</u>	
$5s7s \ ^3S$			1.69	0.30	0.12	
$5s8s \ ^3S$				0.47	0.10	
$5s9s \ ^3S$					0.18	
$5p^2 \ ^3P$		0.007	0.000	0.000	0.000	
$5s4d \ ^3D$	<u>14.9</u> (17) ^a	<u>110</u> (190) ^a , (85) ^b	<u>3.51</u>	<u>0.25</u>	<u>0.050</u>	
$5s5d \ ^3D$			0.60	0.26	0.14	
$5s6d \ ^3D$				0.34	0.16	
$5s7d \ ^3D$					0.16	
$5s4d \ ^1D$	(3.9) ^c					
$\sum A$	<u>21.6</u>	<u>111</u>	<u>6.64</u>	1.77	0.95	present ^d
τ_{theor} (ns)	<u>46.2</u>	<u>9.0</u>	<u>151</u>	565	1.05×10^3	present ^d
	126	12.1	255	518	881	HSCA

^aCorliss and Bozman, Ref. [16].

^bMiller *et al.*, Ref. [13].

^cTable II.

^dUnderlined values involve primarily MQDT and *R*-matrix calculations of transition rates.

TABLE IX. Theoretical transition probabilities (10^6 s^{-1}) and related measurements. Underlining and HSCA as in Table III.

Lower level	Upper level						
	$5s4d^3D$	$5s5d^3D$	$5s6d^3D$	$5s7d^3D$	$5s8d^3D$	$5s9d^3D$	
$5s5p^3P$	<u>4.32</u>	<u>58.3</u> (64) ^h	<u>26.7</u> (30) ^h	<u>14.2</u>	<u>8.51</u>	<u>5.51</u>	
$5s6p^3P$		0.17	3.50	2.20	1.40	0.97	
$4d5p^3P$			0.000	0.000	0.000	0.000	
$5s7p^3P$			0.008	0.91	0.62	0.43	
$5s8p^3P$				0.004	0.27	0.21	
$5s9p^3P$					0.001	0.11	
$4d5p^3F$		0.000	0.000	0.000	0.000	0.000	
$5s4f^3F$			0.13	0.024	0.011	0.006	
$5s5f^3F$				0.083	0.014	0.006	
$5s6f^3F$					0.042	0.006	
$5s7f^3F$						0.021	
$\sum A$	<u>4.32</u>	<u>58.5</u>	<u>30.3</u>	<u>17.4</u>	<u>10.9</u>	<u>7.27</u>	present
τ_{theor} (ns)	<u>231</u>	<u>17.1</u>	<u>33.0</u>	<u>57.5</u>	<u>91.7</u>	<u>138</u>	present
τ_{expt} (ns)		19.0	36.8	62.8	99	143	HSCA
		17.1±0.8	34.0±3.1	74.0±4.8	108±5	232±8	a
		16.0±0.6	32.1±1.6				b
		16.7±1.0					c
		16.4±0.2					d
	$(4.1±0.6) \times 10^3$						e,f
	$(3.4±3) \times 10^3$						g

^aOsherovich *et al.*, Ref. [25].

^bJönsson *et al.*, Ref. [26].

^cBrinkmann, Ref. [33].

^dAndrä *et al.*, Ref. [34].

^eBorisov, Penkin, and Redko, Ref. [40].

^fBorisov, Penkin, and Redko, Ref. [41].

^gMiller *et al.*, Ref. [13].

^hUeda, Ashizawa, and Fukuda, Ref. [36].

TABLE X. Theoretical transition probabilities (10^6 s^{-1}) and related measurements. Underlining and HSCA as in Table III.

Lower level	Upper level							
	$4d5p^3F$	$5s4f^3F$	$5s5f^3F$	$5s6f^3F$	$5s7f^3F$	$5s8f^3F$	$5s9f^3F$	
$5s4d^3D$	<u>24.1</u> (48) ^a , (53) ^b	<u>30.1</u> (19) ^b	<u>19.6</u>	<u>12.4</u>	<u>8.13</u>	<u>5.56</u>	<u>3.95</u>	
$5s5d^3D$		7.62	0.62	0.098	0.20	0.005	0.39	
$5s6d^3D$			1.91	0.36	0.12	0.021	0.12	
$5s7d^3D$				0.65	0.18	0.055	0.039	
$5s8d^3D$					0.27	0.082	0.011	
$5s9d^3D$						0.17	0.001	
$5s10d^3D$?	
$\sum A$	<u>24.1</u>	<u>37.7</u>	<u>22.1</u>	<u>13.5</u> ^c	<u>8.72</u> ^c	<u>5.89</u> ^c	<u>4.51</u> ^c	present
τ_{theor} (ns)	<u>41.5</u>	<u>26.5</u>	<u>45.2</u>	<u>74.0</u> ^c	<u>115</u> ^c	<u>170</u> ^c	<u>222</u> ^c	present
	24.0	50.2	86.7	141	214	269	675	HSCA
	21							a
	19	38						b and HSCA

^aMiller *et al.*, Ref. [13], ±20% estimated uncertainty.

^bCorliss and Bozman, Ref. [16], ±40% estimated uncertainty.

^cTransitions to ³G states are not taken into account.

The eigenstates of Eq. (1) can be calculated using a variety of methods, including close-coupling and configuration-interaction techniques. The most compact and efficient description of the strongly perturbed Rydberg states is obtained using multichannel-quantum-defect theory (MQDT) [6–8]. For many years MQDT has been used as a tool to analyze and interpret such spectra, typically requiring a least-squares fit of the short-range MQDT parameters (i.e., elements of a smooth reaction matrix K) to the experimental spectrum [11]. Recent progress achieved in Refs. [3–6] has demonstrated that these parameters can now be calculated *ab initio* [once the one-electron potential $V(r)$ is known] by a small-scale eigenchannel R -matrix calculation. This is a significant improvement over the semiempirical fitting approach, since the latter tends to be ambiguous and nonunique when applied to systems with more than two or three channels, or to spectra covering a broad energy range.

The final result of the eigenchannel R -matrix calculation, conducted in LS coupling exactly as in Refs. [3–5], is the smooth, short-range reaction matrix K_{ij} and a set of dipole-transition matrix elements. We will prefer in the following discussion to deal with the eigenchannel representation in which K is transformed into diagonal form. That is, we denote the eigenvalues of K by $\tan\pi\mu_\alpha$ and the corresponding orthonormal eigenvectors by $U_{i\alpha}$. An eigenfunction ψ_α , at energy E , of the two-electron Hamiltonian (1) is then represented, beyond the R -matrix reaction-zone radius r_0 , as a close-coupling-type expansion including both open and closed ionization channels:

$$\psi_\alpha = Ar^{-1} \sum_i \phi_i(\Omega) U_{i\alpha} [f_i(r) \cos\pi\mu_\alpha - g_i(r) \sin\pi\mu_\alpha],$$

$$r > r_0. \quad (2)$$

Here A denotes an antisymmetrization operator, while the i th channel function $\phi_i(\Omega)$ includes the full eigenfunction of Sr^+ , LS coupled to the orbital and spin wave functions of the outermost electron of Sr. In Eq. (2) a linear combination of radial Coulombic wave functions (f_i, g_i) , respectively regular and irregular at $r=0$, is used to represent the wave function of the outermost electron at $r > r_0$ where exchange is negligible. At a given total energy E , the energy in the Coulomb functions is $\varepsilon_i = E - E_i$, where E_i is the energy of the ionization threshold of Sr in channel i . In practice the radius r_0 is taken somewhat larger than would be necessary to fit the highest relevant excited state of Sr^+ inside the reaction volume. For the calculations of this paper, involving transitions to the seven lowest states of Sr, $r_0 = 18$ –25 a.u. was used typically, with 100–200 variational basis functions per symmetry.

To describe electric dipole transitions from an initial state ψ_0 of Sr to a final-state wave function ψ_α , we need a set of reduced dipole matrix elements $D_\alpha = \omega^{-1} \langle \psi_\alpha | \nabla_1^{(1)} + \nabla_2^{(1)} | \psi_0 \rangle$. Here ω is the transition frequency in a.u. and $\nabla_i^{(1)}$ represents the irreducible components of the (rank 1) gradient operator, for the i th electron. The set of smoothly energy-dependent short-range parameters $(D_{\alpha, \mu_\alpha}, U_{i\alpha})$ contains all the information

needed to determine the energy levels, photoabsorption oscillator strengths, and other observables. Note that if N channels are retained in the expansion (2), there will be N values of the ionization channel index i and also N values of the eigenchannel index α labeling alternative linearly independent solutions to the Schrödinger equation at energy E .

Oscillator strengths can now be rapidly calculated from the short-range MQDT parameters by “eliminating” closed channels in standard MQDT fashion. For details, see Ref. [10] (pp. 60–63). Velocity values for transition probabilities are given in Tables I–X. These typically differed with length values by $\sim 10\%$, although the discrepancy tends to be larger than this when the transition rate is small.

IV. DISCUSSION

Calculated transition rates (A) for the 1P decays to 1S and 1D states are reported in Tables I and IV. Sums of these A values are given below each column in Table IV, and the inverse is given in the next row as the theoretical lifetime τ_{theor} . The following row gives τ obtained using HSCA values for all transitions, thereby indicating the differences between the MQDT and HSCA values. This is followed by experimental lifetimes, with the reference numbers for these at the right. Tables III, V, and VI give A values and lifetimes for 1S , 1D , and 1F upper states, following the same format as in Table IV. Tables VII–X repeat this for the triplet S , P , D and F states. Finally, in Table XI, we list transitions involving three “parity-unfavored” upper states, i.e., having parity opposite to $(-1)^L$.

The MQDT calculated 1P decays to 1S and 1D states are compared to experimental A values in Table I. The calculated 5^1P – 4^1D f value is too small to be accurate, and the agreement between theory and experiment is within the experimental uncertainty of $\sim 20\%$ for most other cases. The theoretical result is high for transitions from the 6^1P state; this may be due to the close proximity of this state to the 7^3P_1 state, as spin-orbit mixing will lower A for the allowed transition. (Spin-orbit mixing is not included in the calculation.) The excellent agreement for most other transitions is reconfirmed by the lifetime comparisons at the bottom of Table IV. (There the 6^1P state calculation is somewhat closer to the measured lifetimes in Table IV than to the measured f values in Table I.) The MCHF results of Vaecck *et al.* also compare favorably for most of these transitions, as seen in the tables. There we have reported their length result using calculated energies; their other values sometimes are quite different.

As seen in Table V, transitions to the 5^1P state dominate the radiative decay of the $5s5d$, $5p^2$, and $5s6d$ 1D states. The comparison to 1D state lifetime measurements, at the bottom of Table V, thus primarily tests these MQDT calculated values. The agreement is within $\sim 10\%$ for all but the $(5p)^2$ 1D state, where it is 35% different. This discrepancy might be related to neglect of the so-called “dielectronic polarization term,” describing the polarization effect of one electron on the core, which in turn acts on the second valence electron. This term

TABLE XI. HSCA transition probabilities (10^6 s^{-1}) and related measurements.

Lower level	Upper level			
	$4d5p \ ^1D^0$	$5p^2 \ ^3P^e$	$4d5p \ ^3D^0$	
$5s4d \ ^1D$	35.8 (39) ^c ,(32) ^d ,(33) ^e			
$5s5p \ ^3P$		61.0 (120) ^d ,(114) ^j		
$5s6p \ ^3P$		0.000		
$4d5p \ ^3F$		0.000		
$5s4d \ ^3D$			72.3 (116) ^e ,(133) ^d	
$5s5d \ ^3D$			0.000	
$5s^2 \ ^3P^e$			0.002	
$\sum A$	35.8	61.0	72.3	HSCA
τ_{theor} (ns)	27.9	16.4	13.8	HSCA
τ_{expt} (ns)	21.9±1.6			f
		10.2±2.4 ^a		g
		8.3±0.4 ^{a,b}		h
		8.8±1.2 ^a		f
		7.8±1.8 ^b		f
		7.89±0.05 ^b		i
	22	8.3		d

^aFor $5p^2 \ ^3P_1$.^bFor $5p^2 \ ^3P_2$.^cUsing τ_{expt} and Table II branching.^dCorliss and Bozman, Ref. [16].^eMiller *et al.*, Ref. [13].^fBrinkmann, Ref. [33].^gGornik, Ref. [24].^hJönsson *et al.*, Ref. [26].ⁱAndrä *et al.*, Ref. [34].^jUeda, Ashizawa, and Fukuda, Ref. [36].

seems to be most important for states such as $5p^2$, in which both electrons are at comparable radii. For the higher 1D states there is an appreciable or major contribution from transitions to states other than the 5^1P state. The experimental lifetimes, therefore, test a combination of the MQDT and CA values. The agreement between calculated lifetimes and experimental values is very good, the difference being smaller than the disagreement between experiments.

Transitions terminating on the 4^1D state dominate the radiative decay of most 1F states, and measurements of these 1F state lifetimes provide a clear test of these A values from the MQDT calculation. This comparison is made in Table VI, where good (0–20%) agreement occurs with the exception of the $4d5p \ ^1F$ case. The MCHF values of Vaeck *et al.* are also in reasonable agreement with the present MQDT values and with experiment except for this $4d5p \ ^1F$ case. The f value is very small for this exception, implying severe cancellation in the integral and extreme sensitivity of calculations to minor wave function changes. The remaining singlet cases, 1S decays in Table III, are not dominated by the MQDT-calculated transition, and the experimental lifetimes test a combination of MQDT and HSCA values. The agreement is excellent for four cases, 40% for the 9^1S state and ambiguous for the $(5p)^2 \ ^1S$ state due to a large experimental discrepancy. While Ref. [16] does not have as high an accuracy as Ref. [24], the factor of 2.4 difference between these experiments casts doubt on both values for the $(5p)^2 \ ^1S$ state lifetime.

Turning now to the triplet transitions, the one MQDT result in Table VII agrees well with experiment, and the CA values for 7^3S agree well with the only other measurements. We have found no measurements of 3P state lifetimes (Table VIII), but all 3D states in Table IX have been measured. Here the 5^3D – 8^3D cases agree within 20% with the calculated value, the 9^3D state has 40% disagreement, and the 4^3D state has a large fractional disagreement but a small difference in magnitude. The f value of this transition is very small, and fractional agreement is not meaningful. Only CA values have been calculated for the parity-unfavored states in Table XI, for which the parity differs from $(-1)^L$, and as can be seen the agreement with experimental lifetimes varies considerably.

Although no lifetime measurements are available for 3P and 3F states, four of the MQDT values in Tables VIII and X can be compared to measurements from Refs. [13] and [16]. Discrepancies for two cases in Table VIII are within the experimental uncertainty or differences, but discrepancies in Table X are not. In summary, the MQDT calculation typically obtained $\sim 10\%$ differences between length and velocity integrations, and this plus experimental uncertainties bracket most of the discrepancies with experiment. However, much larger discrepancies occur for a small but significant fraction of the transitions.

The remaining A values in Tables III–XI were obtained using the HSCA method. A values for some of these transitions have been reported by Zilitis [42], Gor-

nik [24], and others using the CA with various integral-truncation schemes where needed. Where we have made comparisons, the present HSCA results usually agree with these within 10%. With the exception of the resonance line, the HSCA does quite poorly (typically a factor of 2) on $n^1P_1-5^1S_1$ transitions, as can be seen in the lifetime comparison at the bottom of Table IV. It works equally poorly for lifetimes of the 7^1S , 8^1S , and 9^1S state in Table III, but acceptably (10–25%) for the 10^1S and 11^1S states. On the other hand, HSCA lifetimes for five 1D states in Table V are only 15–40% from experimental values. For the 1F states in Table VI, most HSCA-deduced lifetimes are again in error by about a factor of 2. The remaining case where a full set of HSCA values are experimentally checked is in Table IX, involving the 3D states. Here the HSCA results are quite close to experiment (8–15%) except for the 9^3D state where a 60% discrepancy occurs. Overall, the HSCA gives values of reasonable accuracy for transitions out of the 1D and 3D states, but not from the 1S , 1P , or 1F states. It works about half the time, and its failures are not very predictable.

The combination of experimental transition probabili-

ties in Table I, measured lifetimes in Tables III–XI, and MQDT, MCHF, and HSCA values in Tables I–Xi are the best values currently available for spin-allowed transitions of Sr. In most cases this combination of information appears to achieve net uncertainties of < 30%, but untested HSCA values must be generally regarded as uncertain by about a factor of 2 due to failure to account for configuration mixing. Reference [13] provides some additional experimental checks on transitions to the 5^1P , 5^3P , 4^1D , and 4^3D states, generally confirming this overall conclusion. Some experimental information regarding spin-changing transitions is given in Tables II and VIII, but this spin-orbit mixing was not included in the present calculations.

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*Present address: Van der Waals-Zeeman Lab., Valkenierstrasse 65/67, University of Amsterdam, 1018 XE Amsterdam, The Netherlands.

†Present address: Department of Physics and Astronomy, University of Toledo, Toledo, OH 43606.

‡Quantum Physics Division, National Institute of Standards and Technology.

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