

Lifetimes of alkali-metal—atom Rydberg states

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(Received 9 January 1984)

We report a comprehensive set of calculations of the lifetimes of Rydberg states in alkali-metal atoms, using realistic potentials to represent the atomic core. The effects due to core polarizability, spin-orbit interaction, and blackbody radiation are explicitly included. A complete cross reference listing of all available experimental lifetimes is also attempted. The results compare well with experiment for all principal quantum numbers n examined and should provide guidance for future experimental investigations. The relation of the present approach to other works is discussed.

I. INTRODUCTION

Interest in accurate information on the atomic structure has increased substantially in recent years. In the very basic research branch of quantum optics, the development of new and better lasers and other optical devices places an increasing demand for accurate information on the atomic and molecular states, and particularly on the high-lying Rydberg states.

With the advent of the laser systems we have been able to selectively reach by photon absorption highly excited states. Thus there is a plethora of experimental results which we would like to, and should be able to understand and explain. Detailed and accurate theoretical studies of atomic and molecular Rydberg states will provide information and knowledge that will enable us to predict the feasibility of new "lasing" systems, and will single out the atoms and molecules in which novel processes are most likely to take place and be observed.

With this perspective, the present study was undertaken of transition probabilities and oscillator strengths. The important effects of core polarizability, spin-orbit interaction, and blackbody radiation are explicitly treated. The results are given in the form of the calculated lifetime for each excited state. The lifetime is equal to the inverse of the total transition probability from the state under study to all other accessible states, in this case via a dipole transition. The final result depends, therefore, on a number of transition matrix elements, and accurate theoretical predictions for lifetime values require an accurate description of *all* the states involved.

Experimentally, the lifetime of an excited species can be directly measured or indirectly deduced by a variety of methods each having one or more limitations. Extensive reviews of the experimental approaches to lifetime measurement were given by Corney¹ and by Imhof and Read.²

An atomic electron in an excited (Rydberg) state is occupying an "orbit," the major portion of which is spatially removed from the nucleus and the remaining atomic "core" electrons. Thus it experiences an almost purely Coulombic field. Its energy level is given in Rydberg units by

$$E_{nl} = -\zeta^2 / (n - \mu_l)^2, \quad (1)$$

where ζ is the net charge of the core. This is the classical equation of the Bohr-Sommerfeld model for the atom, apart from the appearance of μ_l , the familiar "quantum defect" of the state under consideration; for hydrogen we have (by definition) $\mu_l = 0$. The quantum defect represents the effect on the electron's motion of all the other (core) electrons. In the limit of the Rydberg series, $n \rightarrow \infty$, the quantum defect and the zero-energy scattering phase shift δ_l are simply related by

$$\pi\mu_l(n \rightarrow \infty) = \delta_l(E=0) \quad (2)$$

which provides an important connection between the discrete and continuum spectra of atoms.³

Systems other than hydrogen having one electron (i.e., ionized atoms) are called "hydrogenlike." For atoms and ions with more than one electron, if the core is compact enough and the principal quantum number N of the outermost electron is sufficiently high, the latter behaves largely as if it were in a pure Coulombic field.

In the absence of external fields, the hydrogenic problem is analytically solvable. The energy levels and the wave functions of the electron are known exactly and all information about the atom (ion) is thus available. The more-than-one-electron problem, however, can be treated only approximately, and in that case our experience from the hydrogenic case serves as a guide in tackling this problem. In the following we outline a short review of previous treatments of Rydberg states and then describe the present approach. Atomic units are used throughout and Rydberg units for energy.

II. PREVIOUS APPROACHES

The traditional way of describing Rydberg-state wave functions and transition probabilities is by the quantum-defect theory which was implemented by Bates and Damgaard in their classic 1949 paper⁴ treating bound-bound state transitions. Burgess and Seaton extended this work to calculating photoionization cross sections in their also often quoted paper.⁵

The basic (approximate) Schrödinger equation that the optical electrons obey is the following:

$$d^2 P_{nl}(r)/dr^2 - \left[V(r) - E_{nl} + \frac{l(l+1)}{r^2} + H(r) \right] P_{nl}(r) = 0, \quad (3)$$

where $V(r)$ is the potential experienced by the optical electron, E_{nl} is the eigenvalue, and $H(r)$ contains all other interactions (predominantly exchange and relativistic effects). For large r we have

$$H(r) \rightarrow 0 \quad \text{as } r \rightarrow \infty, \quad (4)$$

$$V(r) \rightarrow -2(Z-N)/r = -2\xi/r \quad \text{as } r \rightarrow \infty, \quad (5)$$

where Z is the nuclear charge and N is the number of the screening electrons. In this asymptotic case of large distances r the solution of (3) is a Whittaker function and can be given analytically as

$$P_{nl}(r) = (-1)^{n+l+1} \xi^{1/2} K(n^*, l) (2\rho/n^*)^{n^*} \times \exp(-\rho/n^*) \sum_t b_t(n^*, \rho) \rho^{-t}, \quad (6)$$

where

$$\rho = \xi r, \quad (7a)$$

$$K(n^*, l) = [\eta(n^*) n^* \Gamma(n^* + l - 1) \Gamma(n^* - l)]^{-1/2}, \quad (7b)$$

$$b_0 = 1, \quad (7c)$$

$$b_t = (n^*/2t) [l(l+1) - (n^* - t)(n^* - t + 1)] b_{t-1}, \quad (7d)$$

$$(n^*)^2 = -\xi^2/E_{nl} = (n - \mu_l)^2, \quad (7e)$$

and η is a function of n^* close to unity:

$$\eta(n^*) = 1 + [2/n^{*3}] d\mu(\epsilon)/d\epsilon, \quad (8)$$

where ϵ is the electron's energy (in the discrete or continuum spectrum).

The quantity of interest to us is

$$R(nlj; n'l'j') = \int_0^\infty dr P_{nlj}(r) r P_{n'l'j'}(r), \quad (9)$$

and the calculation of the normalized wave functions P_{nlj} is central to the present work. Bates and Damgaard⁴ evaluated $R(nlj; n'l'j')$ by using a series expansion of the form (6) for both ground- and excited-state wave functions and integrating analytically. This yields a double summation which sometimes presents difficulties as pointed out by Burgess and Seaton.⁵

The Bates-Damgaard (BD) method utilizes the experimental binding energy of the active electron to construct an approximate radial wave function which is accurate at large radial distances: a Coulomb wave function with the right behavior at infinity and the right energy. The length form of the electric dipole matrix element, which emphasizes large radial distances, is then evaluated using two such wave functions. The resulting transition probability depends on the initial and final energies and angular momenta of the active electron, but not on the details of the atomic potential at small radii.

The basic postulate of the BD method, also known as the Coulomb approximation (CA), is that nearly all of the dipole length matrix element is contributed by the wave functions at radii where the active electron is outside the

core (the charge distribution of the rest of the electrons and the nucleus); the contribution from smaller radii is either neglected (by using a cutoff radius) or severely approximated (by introducing a modifying factor into the approximate wave function, which prevents the radial integral from diverging at the origin.) The above assumptions are not valid in two cases: (a) when the active electron is in an inner orbital with very small amplitude outside the core or (b) when the outer contribution to the matrix element is very small because of cancellation so that the value of the inner contribution is important. The BD approximation is only valid for electrons not having large wave-function magnitudes at small r ; this criterion may be stated as

$$n^* > l + 1 \quad (10)$$

for valid use of the CA.

Stewart and Rotenberg⁶ extended the CA retaining the Coulomb field for large r , but using a "scaled" Thomas-Fermi potential for small r , i.e., $r < r_0$ where r_0 is the radius of the Thomas-Fermi ion (a function of N/Z only). This approach accounts, to some extent, for the effects of the nonactive atomic electrons and yields a wave function which for $r > r_0$ is identical to the BD one, apart from a normalization factor. To obtain an even more realistic inner potential Warner⁷ proposed the use of the Thomas-Fermi-Dirac model⁸ to account for the electron exchange effects. In all the above approaches experimental (spectroscopic) eigenvalues have been used, eliminating the need of a self-consistent calculation.

Heavens⁹ has used the tables produced by Bates and Damgaard⁴ to calculate the radiative transition probabilities from and the lifetimes of the lower Rydberg states of Li, Na, K, Rb, and Cs, and his values are among the most quoted among the CA ones in the literature. Various research groups have been recalculating transition probabilities and radiative lifetimes in the CA, implementing various series cutoff criteria. Comparison to their work will be made below.

Anderson and Zilitis¹⁰ performed extensive calculations and tabulations of the alkali-metal-atom optical oscillator strengths using a realistic potential throughout. They used an atomic potential of the Hartree type and integrated the Schrödinger equation employing spectroscopic eigenvalues. The wave functions at large r were described by asymptotic series representing the solutions in a Coulomb field.

An alternative approach, the so-called "numerical Coulomb approximation" (NCA) method, has been introduced and applied by Lindgaard and Nielsen.¹¹ In this approximation, the wave functions are obtained numerically by direct *inward* integration of Schrödinger's equation starting with the correct asymptotic boundary conditions. The integration is terminated at a certain small distance r_c , so that the wave function is normalized to unity, and the obtained expectation value $\langle r \rangle$ agrees with the hydrogenic formula

$$\langle r \rangle = \int_0^\infty dr r P_j^2(r) = \frac{1}{2\xi} [3n_j^{*2} - l(l+1)], \quad (11)$$

where the effective quantum number n^* replaces the integer hydrogenic value n .

The Bates-Damgaard approximation has a sound basis when treating transitions between states with medium to high quantum numbers n , and especially between "non-penetrating" states; the numerical results verify this. Whenever one of the principal quantum numbers n is low, however, especially when $n^* < l + 1$, the state is not described accurately and this can lead to significant discrepancies between theory and experiment (as we will see below).

III. THE PRESENT APPROACH

A. The potential

The major problem and error of the BD method is in approximating the atomic potential within the core region by its asymptotic value $-2\xi/r$. To date, several attempts have been made to rectify this shortcoming, by assuming certain analytical or numerical approximations of the core part of the atomic potential.^{6,7,10,11}

The present approach is aimed at (i) utilizing the fact that the optical electron is experiencing for most of the time a basically hydrogenic (Coulombic) field; (ii) accounting in a realistic way for the potential within the core; and (iii) accounting for the major additional electron-core interactions, notably the core polarizability and spin-orbit interaction effects. The atomic potential used in Schrödinger's equation within the present treatment is

$$V(r) = V_c(r) + V_p(r) + V_{s.o.}(r). \quad (12)$$

It consists of three terms.

(i) The single-electron central field V_c due to the nucleus and the core electrons. For Li, Na, and K it is constructed from analytic Hartree-Fock orbitals¹² of the occupied shell electrons, as

$$V_c(r) = V_{HF}(r) = -(Z - N)/r - 2 \sum_{\alpha} (2l_{\alpha} + 1) \xi_{\alpha}(r)/r, \quad (13)$$

$$\xi_{\alpha}(r) = \int_r^{\infty} dx (1 - r/x) P_{\alpha}^2(x), \quad (14)$$

where Z is the nuclear charge, N is the number of electrons in the closed shells, and $P_{\alpha}(r)$ is the Hartree-Fock orbital in the α th shell. In the case of Rb and Cs the appropriate Hartree potentials¹³ were used.

(ii) The polarization potential V_p which represents the effect of the induced core electron moments on the active electron, and can be taken to be of various forms: e.g., (a)

$$\begin{aligned} V_p(r) = & -\frac{1}{2} \alpha_d r^{-4} \{1 - \exp[-(r/r_1)^6]\} \\ & -\frac{1}{2} \lambda r^{-6} \{1 - \exp[-(r/r_1)^8]\} \\ & + (c_0 + c_1 r) \exp(-r/r_0), \end{aligned} \quad (15)$$

where the parameters α_d , λ , r_0 , r_1 , c_0 , and c_1 are given by Weisheit¹⁴ for Na, K, Rb, and Cs, and by Bottcher¹⁵ for Li; or (b) the form used by Norcross,¹⁶

$$\begin{aligned} V_p(r) = & -\frac{1}{2} \alpha_d r^{-4} \{1 - \exp[-(r/r_c)^6]\} \\ & -\frac{1}{2} (\alpha_q - 6\beta a_0) r^{-6} \{1 - \exp[-(r/r_c)^{10}]\} \end{aligned} \quad (16)$$

which has only one adjustable parameter. Here α_d and α_q are the static dipole and quadrupole core polarizabilities, respectively, and β is a dynamic correction to the former, representing the time delay in the core-dipole response to the active electron's motion. All three quantities can be calculated *ab initio*.¹⁷ It should be mentioned that the values for the quadrupole polarizability inferred from experiment seem to agree poorly with the calculated ones.¹⁷ The parameter r_c is obtained by fitting the observed few lowest-energy levels of the active electron. The advantage of the model potential used by Norcross¹⁶ over similar approaches is that one has only a single parameter to fit to experimental data and therefore can be more realistic.

(iii) The spin-orbit interaction potential $V_{s.o.}$ is taken to be the term in the Pauli equation. The full relativistic form is used to ensure that the potential has the correct behavior near the origin, and is given by

$$V_{s.o.}(r) = \frac{1}{2} \alpha^2 f(r)^2 (1/r) [dV_m(r)/dr] \vec{L} \cdot \vec{S} \quad (17)$$

with

$$f(r) = \{1 + (\alpha^2/4)[E - V_m(r)]\}^{-1}, \quad (18)$$

where $V_m(r)$ is the model potential consisting of the core plus the polarization potentials, defined above:

$$V_m(r) = V_c(r) + V_p(r). \quad (19)$$

This model potential has the following limiting values:

$$V_m(r=0) = -2Z/r, \quad (20)$$

$$V_m(r \rightarrow \infty) = -2(Z - N)/r. \quad (21)$$

Away from the origin $f(r)$ is equal to unity; this is not true near $r=0$, however, and its full expression is retained so that the series expansion of the wave function there has the correct behavior.

All three potential terms are known exactly for distances r larger than the atomic core radius, or at least for r larger than a few times that. This is true because the central atomic field becomes always $-2(Z - N)/r$ for $r > r_c$. Therefore, whatever approximations are involved in constructing the model potential, these approximations and their effects are limited to distances less than the atomic core radius. This radius is not strictly defined; in any case, it is of the order of only a few Bohr radii and does not differ from model to model by more than 10–20%.

Electron exchange has not been *explicitly* included above. Its effects are expected to be small. Electron exchange, as well as relativistic terms in the Pauli approximation, other than the spin-orbit interaction, are *implicitly* included, in a local-potential form, through the term

$$(c_0 + c_1 r) \exp(-r/r_0)$$

of the polarization potential. Nonlocal potential terms would greatly increase the amount of computational work. The effect of spin-orbit interaction is treated here explicitly because it describes the splitting of the various

levels according to the total angular momentum j , whereas the other terms of the Pauli approximation move the center of gravity of these levels. This center of gravity of several terms was fitted in obtaining the polarization potential parameters of Eqs. (15) and (16) above.

B. The wave functions

The traditional self-consistent-field calculation of wave functions entails outward integration of the appropriate Schrödinger equation, starting at $r=0$, and inward integration from the practical infinity. The inward integration is continued until the outer classical turning point where the solution is matched to the outward one. The energy is adjusted so that the correct number of nodes is present and the wave function is smooth at the matching point. However, even if the best energy eigenvalue within a model is obtained, this does not necessarily imply the best or, at least the most correct wave function possible. This is true because in all cases the atomic potential can be described only *approximately*.

In the present approach, the Rydberg-state radial wave functions are obtained by direct *inward* integration of the appropriate Schrödinger equations. After establishing the model potential $V_c + V_p + V_{s.o.}$ we employ the experimental energy level values and integrate Schrödinger's equation inwards with the correct boundary condition at infinity, in the same way as Lindgaard and Nielsen.¹¹ Since we use an accurate potential, however, the wave functions so obtained are accurate also at and near the origin, something we have tested for several typical cases. This is ensured by matching the inward integration with the outward one at the *inner* classical turning point, i.e., near the origin. Since we do not iterate the process, this numerical wave function may have a discontinuous first derivative at the matching point. The latter's location, however, is very close to the origin and has no noticeable effect on the transition matrix elements.

C. The transition probabilities

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

$$A(\gamma J \rightarrow \gamma' J') = \frac{4}{3} \alpha^2 \left[\frac{\Delta E}{R} \right]^3 \frac{S(\gamma J, \gamma' J')}{2J+1} \quad (22)$$

and

$$f(\gamma' J' \rightarrow \gamma J) = \frac{1}{3} \frac{\Delta E}{R} \frac{S(\gamma J, \gamma' J')}{2J'+1}, \quad (23)$$

respectively, where

$$S(\gamma J, \gamma' J') = |(\gamma S L J || \vec{e} \cdot \vec{r} || \gamma' S' L' J')|^2 \quad (24)$$

is the line strength, α 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 line strength takes the explicit form:

$$S(nl j \rightarrow n' l' j') = (2j+1)(2j'+1) \left\{ \begin{matrix} 1 & l & l' \\ \frac{1}{2} & j' & j \end{matrix} \right\}^2 \max(l, l') \times R(nl j; n' l' j') \quad (25)$$

with

$$R(nl j; n' l' j') = (nl j || r || n' l' j')$$

[cf. Eq. (9)].

A derivation of the dipole transition moment $R(nl j; n' l' j')$ including, to a first-order correction and in a consistent way, the effects of core polarization by the valence electron leads to the replacement^{18,19}

$$r \rightarrow r [1 - \alpha_d(\omega) \kappa(r) / r^3] \quad (26)$$

in the integral (9). Here $\alpha_d(\omega)$ is the dynamic dipole polarizability of the core at the transition frequency $\omega = \Delta E$, and $\kappa(r)$ is the ratio of the true induced dipole moment operator to its asymptotic form at large r . For discrete transitions $\alpha_d(\omega)$ is usually taken to be equal to the static value $\alpha_d(0) = \alpha_d$, which is a good approximation for the lower transitions. A better first approximation is²⁰

$$\alpha_d(\omega) \approx \alpha_d(0) / [1 - (\omega / \Delta E_{rc})^2] \quad (27)$$

where ΔE_{rc} is the resonance energy of the core; this approximation is only valid for frequencies less than this energy. The ratio $\kappa(r)$ is traditionally modeled either as

$$\kappa(r) = 1 - \exp[-(r/r_c)^m], \quad (28)$$

where $m=3$ may be the most physically appropriate choice,^{18,19} or^{20,21}

$$\kappa(r) = r^3 / (r^2 + r_c^2)^2 \quad (29)$$

which at $r \rightarrow 0$ corresponds to $m=4$ in (28). The effective core radius r_c is usually taken either so that the theoretical oscillator strengths reproduce characteristic features of the experimental oscillator strengths and/or photoionization cross sections, or in some other semi-empirical way.

A more recent treatment by Moore and co-workers²² calculates the cutoff functional in a self-consistent way and may be a more desirable choice. They use a Hartree-Slater central potential to describe the alkali-metal-atom core, and for the valence electron the core polarization potential

$$V_p(r) = -\frac{\alpha_d}{2r^4} \frac{P^2(r)}{1 - 4\pi\alpha_d \rho_m(r) / Z_m} \quad (30)$$

with

$$Z_m \rho_m(r) = [1 - \exp(-\lambda r^p / a_0^p)] n(r), \quad (31a)$$

$$\int \rho_m(r) dr = 1, \quad (31b)$$

$$P(r) = \int_0^r \rho_m(r') dr', \quad (31c)$$

where the parameters p , λ , and α_d are inferred from experiment, $n(r)$ is the total ion core electron density, and $P(r)$ is exactly equal to the ratio $\kappa(r)$ in Eq. (26).

D. The blackbody-radiation (BBR) effect

The population of the higher states can be significantly affected by the blackbody radiation in which the atoms are embedded.²³ If there exist other states that are energetically very close to the one under study, a significant depletion of its population occurs by stimulated emission and absorption. Specifically, the total transition probability A_m from the m th state to "anywhere" is generally obtained from²⁴

$$\frac{g_m}{\tau_m^*} = g_m A_m = \sum_{m' < m} g_{m'} A(m \rightarrow m') (\langle n \rangle + 1) + \sum_{m'' > m} g_{m''} A(m'' \rightarrow m) \langle n \rangle, \quad (32)$$

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 :

$$\langle n(h\nu) \rangle = \frac{1}{e^{h\nu/k_B T} - 1} = \frac{1}{e^{|\Delta E|/k_B T} - 1}, \quad (33)$$

g_m is the degeneracy of the m th state, and τ_m^* its effective lifetime. For low-lying states this effect is negligible because $\Delta E \gg k_B T$ at temperatures below 1000°C.

IV. COMPUTATIONAL APPROACH

To obtain the radial wave functions of the active electron in its initial and final states we have to solve the second-order differential equation:

$$d^2 P_i / dr^2 = [V_i(r) - E_i] P_i(r), \quad (34)$$

$$V_i(r) = V_c + V_{s.o.} + V_p, \quad (35)$$

where $V_i(r)$ is the sum of the (central) core potential, the spin-orbit interaction potential, and the polarization potential defined earlier. As mentioned above, $E_i = E_{nlj}$ is input here and no iteration is implemented in the solution of (34). This equation is amenable to a solution using the efficient Numerov method. Numerov's approach is more efficient when an equidistant mesh is used for the independent variable. The nature of the potential $V(r)$ which has a pole at the origin ($r=0$) implies the particular need of dense integration (mesh) points near the origin. At intermediate to large distances the mesh points may be considerably less dense, for usual cases. Since, however, our approach is intended for states with typical effective principal quantum numbers $n^* = 5-15$, i.e., with the electron spending most of its time at distances 38-340 a.u., there are limitations to the maximum distance between adjacent mesh points at large r . A new variable taking into account the above two requirements is obtained by the following transformation, proposed by Bratsev²⁵ and implemented by Chernysheva *et al.*²⁶ for cases similar to ours:

$$x = \alpha r + \beta \ln r, \quad (36)$$

where α and β are constants. The variable x behaves like $\ln r$ at small r (a variable employed almost invariably for ground-state self-consistent-field calculations) and like αr

at $r \rightarrow \infty$, which is suitable for spatially extended excited states.

In principle, the new variable takes values in the interval $(-\infty, +\infty)$. For the actual calculations we restrict it to the interval $[x_0, x_N]$ where

$$x_0 = -\beta(10 + \ln Z) \quad (37)$$

corresponding to

$$r_0 \approx 4.5 \times 10^{-5} / Z. \quad (38)$$

An equidistant mesh is used for x ,

$$x_i = x_0 + ih, \quad i = 0, 1, 2, \dots, N \quad (39)$$

and the coefficient α is given by the relation

$$\alpha = (x_N - \beta \ln r_{\max}) / r_{\max}. \quad (40)$$

Finally, the maximum distance r_{\max} considered, the mesh h , the number of mesh points N , and the coefficient β are input parameters.

After introducing the new variable, Eq. (34) yields an equation including a first derivative. To implement Numerov's integration method, this derivative is removed by the transformation

$$y_i(x) = [(ar + \beta)/r]^{1/2} P_i(r) \quad (41)$$

which yields

$$d^2 y / dx^2 = p(x) y, \quad (42)$$

$$p(x) = (\alpha r + \beta)^{-2} r^2 [V_i(r) - E_i] + \beta(\alpha r + \beta/4) / (\alpha r + \beta)^4. \quad (43)$$

The last two equations are solved in a convenient way by the method described in the Appendix.

V. RESULTS AND DISCUSSION

A. General

The approach outlined in Secs. III and IV above was applied to the calculation of lifetimes of s , p , d , and f states with principal quantum n up to at least $n=16$ in the stable alkali-metal atoms, i.e., Li, Na, K, Rb, and Cs. The results are given below in Tables II-VI, and will be discussed separately for each atom. The agreement is excellent for the lower states where the analytic and numerical CA methods are least accurate. The lifetimes shown there depend on the transition probabilities to all dipole-allowed lower states, and the composite final result is affected by the inadequacies of an approximation, like BD, to various degrees, depending on the upper state under consideration. We are impressed, however, at how good NCA¹¹ is overall. This should be contrasted to the fact that the actual expectation value $\langle r \rangle$ of the lowest states is *not* equal to the hydrogenic prediction of Eq. (11) with n^* . Table I shows a comparison between the hydrogenic values and the values obtained numerically by the present approach, as well as by the Hartree-Slater (HS) and Hartree-Kohn-Sham (HKS) predictions.²⁷ This table also indicates the difficulty of a self-consistent-field calculation to converge to a correct result for some excited states.

Higher angular momentum states have negligible quantum defects, since they do not penetrate the atomic core,

TABLE I. Expectation values $\langle r \rangle$.

	$nl \quad j$	Hydrogenic	This work	HS	HKS
Li	$2s \quad \frac{1}{2}$	3.815	3.785	3.755	3.920
	$2p \quad \frac{1}{2}, \frac{3}{2}$	4.694	4.758	4.694	4.814
Na	$3s \quad \frac{1}{2}$	4.021	3.971	4.021	4.277
	$3p \quad \frac{1}{2}$	5.847	5.720	5.690	5.972
	$3p \quad \frac{3}{2}$	5.853	5.725		
	$3d \quad \frac{3}{2}, \frac{5}{2}$	10.387	10.408	10.362	10.425
K	$4s \quad \frac{1}{2}$	4.609	4.702	4.968	5.356
	$4p \quad \frac{1}{2}$	6.436	6.474		
	$4p \quad \frac{3}{2}$	6.462	6.493	6.670	7.088
	$3d \quad \frac{3}{2}$	7.993	9.217		
	$3d \quad \frac{5}{2}$	7.989	9.215	6.261	8.975
	$4f \quad \frac{5}{2}, \frac{7}{2}$	17.874	17.918		
	$4d \quad \frac{3}{2}$	17.536	18.625	17.977	17.986
	$4d \quad \frac{5}{2}$	17.532	18.622		
Rb	$5s \quad \frac{1}{2}$	4.917	4.886	5.292	5.751
	$5p \quad \frac{1}{2}$	6.887	6.797		
	$5p \quad \frac{3}{2}$	6.896	6.886	7.149	7.618
	$4d \quad \frac{3}{2}$	7.640	8.483		
	$4d \quad \frac{5}{2}$	7.642	8.482	6.417	8.794
	$4f \quad \frac{5}{2}, \frac{7}{2}$	17.757	17.860	17.946	17.969
	$5d \quad \frac{3}{2}$	17.157	17.601		
	$5d \quad \frac{7}{2}$	17.166	17.608	12.956	16.185
Cs	$6s \quad \frac{1}{2}$	5.304	5.241	5.886	6.431
	$6p \quad \frac{1}{2}$	7.181	7.137		
	$6p \quad \frac{3}{2}$	7.424	7.367	7.743	8.284
	$5d \quad \frac{3}{2}$	6.059	6.736		
	$5d \quad \frac{5}{2}$	6.118	6.792	5.428	7.930
	$4f \quad \frac{5}{2}, \frac{7}{2}$	17.541	17.738	17.077	

and they decay to other also nonpenetrating states. Thus, hydrogenic matrix elements and transition probabilities can be used; they are given by Green *et al.*²⁸ We must include, however, the effects of the core polarizability which can be simply done in this case, as described by Curtis,²⁹ by the substitution

$$\langle nl || r || n'l' \rangle \rightarrow \langle nl || r || n'l' \rangle \times \left[1 - \frac{\alpha_d / \alpha_0^3}{45^2 (R \lambda_{nl, n'l'})^2} \right], \quad (44)$$

and a cutoff distance is *not* necessary.

1. Effects of spin-orbit interaction

The spin-orbit interaction is a relativistic effect, is proportional to $\langle r^{-3} \rangle$, and becomes more important for the heavier alkali-metals. In lithium it hardly splits the $j = \frac{1}{2}$ and $\frac{3}{2}$ levels in classical spectroscopic studies, whereas for cesium these splittings are substantial and the two j states *must* be treated separately. In terms of the Schrödinger equation and the electron wave function, the spin-orbit interaction can significantly alter the wave function's

TABLE II. Lifetimes of Li (in nsec).

State	0 K	This work 620 K	720 K	Experimental	Other theoretical NCA ^a	Other
3s	30.04	30.04	30.03		30.32	30.0°
4s	56.29	56.04	55.87	52.7(1.6) ^a 55.8(2.8) ^b 48.2(2) ^c 56(1.7) ^d	56.65	57.2° 54.5 ^p 56.9 ^q
5s	102.46	100.43	99.60	102.5(4.0) ^a 113(6) ^b 99(9) ^d	103.0	103° 104.0 ^q
6s	172.44	164.71	162.39	166.4(8.1) ^a	173.2	175.1 ^q
7s	271.06	251.36	246.64	243(17) ^a	272.9	
8s	403.30	362.89	354.55	387(31) ^a	405.3	
9s	574.76	501.79	488.27		578.6	
10s	783.44	666.46	646.39		788.0	
11s	1057.11	869.49	839.71		1061.4	
12s	1369.49	1097.54	1056.70		1374.1	
13s	1745.77	1361.38	1306.57			
14s	2186.09	1660.44	1588.88			
15s	2695.43	1993.76	1902.30			
16s	3278.95	2363.74	2249.15			
17s	3941.81	2771.76	2630.62			
18s	4689.03	3218.25	3046.95			
19s	5525.69	3704.88*	3499.67*			
20s	6456.82	4239.92*	3997.17*			
2p	27.24	27.24	27.24	26.9(0.8) ^a 25.0(1) ^c 26.2(1) ^e 31.9(1.9) ^b 27.2(0.4) ^f 26.4(8) ^g 27.29(4) ^h	27.32	27.2° 26.9°
3p	212.18	211.92	211.78	203(8) ^g 202 ⁱ 235 ^b 182(6) ^j	216.4	209°
4p	391.22	383.94	378.90		402.6	364°
5p	610.25	567.63	550.19		627.8	107°
6p	913.49	792.40	757.50		940.0	
7p	1319.45	1064.50	1007.97		1358.9	
8p	1848.24	1389.97	1307.16		1856.1	
9p	2509.54	1769.69	1656.22		2587.5	
10p	3328.00	2205.04	2055.05		3360.4	
11p	4310.12	2715.92	2523.89		3448.9	
12p	5490.30	3293.97	3052.38		5663.9	
13p	6856.38	3909.02	3611.23			
14p	8270.24	4575.10	4222.49			
15p	10276.38	5436.55	5005.24			
16p	12355.00	6357.23	5846.45			
17p	14702.03	7425.06	6824.89			
18p	17334.06	8721.70	8020.28			
19p	20267.71	10500.14	9681.98			
20p	23518.79	14700.59*	13769.09*			

TABLE II. (Continued).

State	0 K	This work 620 K	720 K	Experimental	Other theoretical NCA ^a	Other
3d	14.64	14.64	14.64	14.9(0.5) ^a	14.66	14.5°
				14.5(0.7) ^k		14.0 ^q
				13.9(0.5) ^e		14.0 ^r
				15.0(1.0) ^c		
				14.0(0.7) ^b		
				16.7(1.0) ^l		
4d	33.49	33.47	33.45	14.60(13) ^m	32.42	35.2°
				32.5(0.9) ^a		32.5 ^p
				31.0(1) ^k		33.5 ^q
				33.0(1) ^e		32.6 ^r
				39.2(2) ^b		
				42.3(1.5) ^l		
5d	63.89	63.64	63.47	61.5(2.9) ^a	63.70	64.8 ^q
6d	108.63	107.34	106.76	103.2(4.0) ^a	108.2	
7d	170.53	166.64	165.31	171.2(8.9) ^a	169.9	
8d	252.39	243.56	241.05	229(14) ^a	251.2	
9d	357.02	340.14	335.95	316(25) ^a	355.9	
10d	487.25	458.29	451.80	436(44) ^a	485.9	
11d	656.66	609.50	599.68	575(56) ^a	648.2	
12d	834.96	765.81	752.38		833.1	
13d	1059.72	959.97	941.58			
14d	1320.55	1184.02	1159.92			
15d	1621.12	1440.69	1409.94			
16d	1964.20	1732.76	1694.41			
17d	2352.63	2065.73	2019.10			
18d	2789.27	2448.44	2393.53			
19d	3276.83	2901.73	2840.57			
20d	3818.19	3506.67*	3450.96*			
4f	72.34	72.24	72.12		72.39	
5f	139.85	138.27	137.21		139.9	
6f	239.04	231.03	227.50		239.1	
7f	375.93	352.96	345.46		376.3	
8f	556.59	506.94	493.71		556.1	
9f	787.11	695.99	675.16		787.6	
10f	1073.59	922.40	891.70		1073.5	
11f	1422.17	1190.20	1147.28		1419.7	
12f	1840.29	1503.83	1445.97		1841.5	
13f	2331.03	1855.33	1778.97			
14f	2902.44	2262.79	2165.56			
15f	3560.47	2726.83	2605.62			
16f	4311.22	3255.17	3106.88			
17f	5160.82	3860.86	3682.68			
18f	6115.42	4569.77	4359.89			
19f	7181.14	5447.83	4208.03			
20f	8363.92	6768.67*	6521.05*			

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TABLE II. (Continued).

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behavior at small r , especially its slope, which also depends on the energy. Thus in the traditional *outward* integration approach a possible error near the origin propagates in an unpredictable way towards large r . This is especially true if a non-self-consistent method is used, i.e., when the energy parameter is kept constant. In the present method the integration is performed *inwards* using the experimentally available energy levels, and the analytically known asymptotic boundary conditions⁴ [cf. Eq. (6)]. Therefore, the wave function is *accurately described* for most of the distances, *independent* of what potential is used to describe the core and polarization effects. The latter normally extend to no more than 10 a.u. whereas the majority of the states studied have an expectation value $\langle r \rangle$ well beyond this number. In the low- r region we still use a very accurate potential, obtained by fitting experimental energies, or, in the worst case, by a Hartree-Slater potential, which for all *practical* purposes is a very good approximation to the "exact" potential. Still, *any* departure from the "exactness" consistent with the experimental energy will cause the wave function to miss passing through the origin, though the latter is a physical requirement. We avoid this difficulty by matching the inward integration with the outward integration, from the origin, at the innermost classical turning point (cf. Sec. III B).

Exactly because of the inward nature of our approach, the spin-orbit interaction potential, being mainly of short range, has minimal effects on the obtained results. This should also be true for the similar approach by Lindgaard and Nielsen.¹¹ Nevertheless, effects of the spin-orbit interaction are inherently included into the treatment through the experimental energies employed, which depend on it directly.

2. Effects of core polarization

As discussed in Sec. III, core polarization affects the calculation of transition matrix elements, which is central to our work, in two ways: first, it changes the wave functions and their energies themselves, and, second, it changes the effective transition operator. The latter is usually more important. In the actual calculations we used the parametric potentials given by Bottcher¹⁵ for Li and by Weisheit¹⁴ for Na, K, Rb, and Cs because they were readily available. Due to the nature of our treatment, the polarization potential, with leading term $1/r^4$, has a small effect on the wave function. We found, how-

ever, that the change in the dipole operator was significant. We investigated the two approaches to the cutoff function $\kappa(r)$ [cf. Eq. (26)], i.e., Eqs. (27) and (29), for the lithium atom. They give different results when applied with the same value for the cutoff radius; the results become comparable, however, when the Hartree-Slater-potential core radius²⁷ is used in conjunction with (27), and the expectation value $\langle r \rangle$ of the outermost occupied orbital (of the core) is used in (28). There has been some controversy in the literature about the arbitrariness of choice for the cutoff radius r_c . We feel that the choice of r_c to be the Hartree-Slater, or Dirac-Slater for heavier atoms, core radius of the atom, is unique as well as physical enough and we have implemented it for Li. Our findings concerning the use of $\alpha_d(\omega)$ rather than $\alpha_d(0)$ are that the results are only slightly different, as expected for the transitions of our interest.

3. Effects of blackbody radiation

Since we are dealing with the alkali metals, temperatures significantly higher than room temperature (300 K) are needed to produce enough metal vapor so that a sufficient experimental signal-to-background ratio can be obtained. A review of all the experimental investigations shows that the temperatures at which data were obtained ranged from 620 to 720 K for Li, to at least 350 K for Cs, and one work was at 5 K. In the following tables we list the lifetimes calculated at absolute zero, the standard reference point, and at two other temperatures, at least one of which is the most commonly employed in the respective experimental studies. The temperatures are well in excess of the room temperature at which the pioneering work of Gallagher and Cooke²³ was performed, and show that the population depletion progresses *rapidly* with the increase of temperature, i.e., effects more drastic than those reported in Ref. 23 are seen even at lower Rydberg states with $n \approx 15$. In our calculations we routinely included contributions from the blackbody-radiation-induced transitions to all dipole-accessible lower states and to 15–20 higher states. The number of higher states was so determined only for numerical convenience, and we made sure that contributions from further higher states were less than 1%. This number should be increased at higher temperatures as well as for higher principal quantum number n . Actually, for the highest levels considered, inclusion of all states that practically contribute to the BBR depletion of population becomes prohibi-

TABLE III. Lifetimes of Na (in nsec).

State <i>nl j</i>	0 K	This work		Experimental	Other theoretical	
		410 K	600 K		NCA ^s	Other
4s $\frac{1}{2}$	37.71	37.71	37.71		39.54	40.5 ^t 88.94 ^u 37 ^v
5s $\frac{1}{2}$	77.64	77.60	77.30	71(5) ^a	80.27	84 ^t 80 ^v 79.3 ^w
6s $\frac{1}{2}$	148.12	146.63	143.21		152.5	159.4 ^x 160 ^v 151.2 ^w
7s $\frac{1}{2}$	256.34	246.88	235.82	269(10) ^a 276(14) ^b	263.2	278.9 ^x 261.8 ^w 280 ^v
8s $\frac{1}{2}$	410.31	381.21	358.10	393(90) ^a 465(40) ^b	422.1	451.9 ^x 418.2 ^w 450 ^v
9s $\frac{1}{2}$	618.17	553.46	512.89	618(43) ^a 713(76) ^b	634.6	683.9 ^x 633.7 ^w 690 ^v
10s $\frac{1}{2}$	888.23	767.22	702.80	1024(49) ^b	913.5	909 ^w 900 ^v 1000 ^v
11s $\frac{1}{2}$	1228.78	1025.35	929.49	1280(130) ^c	1262.8	1250 ^x 1400 ^v
12s $\frac{1}{2}$	1647.63	1330.42	1194.74		1690.0	1900 ^v
13s $\frac{1}{2}$	2152.78	1684.78	1499.98	2270(170) ^b		2180 ^x 2004 ^w 2300 ^v
14s $\frac{1}{2}$	2753.05	2091.20	1847.11			
15s $\frac{1}{2}$	3456.34	2552.58	2238.57			
16s $\frac{1}{2}$	4270.55	3071.52	2676.16			
17s $\frac{1}{2}$	5204.35	3651.16	3162.37			
18s $\frac{1}{2}$	6265.83	4299.35	3704.87			
19s $\frac{1}{2}$	7462.40	5030.01*	4317.94*	7420(170) ^d		7580 ^d
20s $\frac{1}{2}$	8804.24	5901.01*	5062.76*	8900(400) ^d		8940 ^d
21s $\frac{1}{2}$	10 295.91	7908.27*	7061.79*	11 300(600) ^d		10 500 ^d
3p $\frac{1}{2}$	16.14	16.14	16.14	16.40(3) ^e 17(2) ^f 17(2) ^g 17.3(1.0) ^h 14.0(2) ⁱ 15.9 ^j 16.0(2) ^k		17.0 ^t

TABLE III. (Continued).

State <i>nl j</i>	This work			Experimental	Other theoretical		
	0 K	410 K	600 K		NCA ^s	Other	
$3p \frac{3}{2}$	16.09	16.09	16.09	17(2) ^f	16.49	16.9 ^t	
				17(2) ^g			17 ^v
				17.3(1.0) ^h			
				14.0(2) ⁱ			
				15.9 ^j			
				16.0(5) ^l			
				16.1(2) ^m			
				16.0(3) ⁿ			
				16.12(22) ^o			
$4p \frac{1}{2}$	107.19	107.15	107.03	99.5(3) ^p	101.8	106 ^t	
				125(10) ^b			83.15 ^u
$4p \frac{3}{2}$	106.45	106.41	106.29	98(3) ^p		103 ^w	
$5p \frac{1}{2}$	369.86	366.71	357.69	345(43) ^q	342.1	353 ^t	
							351 ^w
$5p \frac{3}{2}$	366.36	363.25	354.38			330 ^v	
$6p \frac{1}{2}$	918.90	860.65	773.57	890(90) ^q	836.7	864 ^w	
							770 ^v
$6p \frac{3}{2}$	908.69	851.25	763.99				
$7p \frac{1}{2}$	1866.17	1463.27	1163.05	1450(100) ^q		1750 ^w	
							1500 ^v
$7p \frac{3}{2}$	1843.52	1511.42	1244.25		1676.0		
$8p \frac{1}{2}$	3333.76	2112.26	1582.58			2600 ^v	
$8p \frac{3}{2}$	3270.98	2257.86	1755.43		2951.0		
$9p \frac{1}{2}$	5059.11	2642.85	1932.53			4100 ^v	
$9p \frac{3}{2}$	5136.77	3007.34	2271.90		4688.0		
$10p \frac{1}{2}$	7264.74	3203.44	2314.43			6000 ^v	
$10p \frac{3}{2}$	8076.90	3956.74	2920.86		7014.3		
$11p \frac{1}{2}$	10025.73	3799.36	2724.06			8500 ^v	
$11p \frac{3}{2}$	11388.64	4866.25	3563.61		10184		
$12p \frac{1}{2}$	13407.10	4417.46	3149.04			1200 ^v	
$12p \frac{3}{2}$	15639.35	5895.48	4292.39		13812		
$13p \frac{1}{2}$	17472.88	5046.97	3583.22			1500 ^v	
$13p \frac{3}{2}$	20791.89	7020.15	5091.57				
$14p \frac{1}{2}$	22286.49	7598.35	5534.95				
$14p \frac{3}{2}$	26918.84	8242.55	5960.94				

TABLE III. (Continued).

State <i>nl j</i>	0 K	This work 410 K	600 K	Experimental	Other theoretical NCA ^s	Other
15p $\frac{1}{2}$	27 844.92	8721.48	6345.72			
15p $\frac{3}{2}$	34 096.89	9567.34	6902.77			
16p $\frac{1}{2}$	34 817.45	10 291.80	7492.36			
16p $\frac{3}{2}$	43 033.69	11 047.54	7946.34			
17p $\frac{1}{2}$	42 821.85	12 043.49	8775.12			
17p $\frac{3}{2}$	51 891.45	12 760.20	9198.90			
18p $\frac{1}{2}$	51 922.58	14 113.02	10 300.47			
18p $\frac{3}{2}$	62 674.89	14 879.12	10 748.14			
19p $\frac{1}{2}$	62 186.16	16 779.35*	12 289.06*			
19p $\frac{3}{2}$	74 843.01	17 648.79	12 794.90			
20p $\frac{1}{2}$	73 653.29	21 033.13*	15 538.60*			
20p $\frac{3}{2}$	89 736.83	22 263.54*	16 261.30*			
3d $\frac{3}{2}$	19.44	19.44	19.44			20.2 ^t 21 ^v
3d $\frac{5}{2}$	19.47	19.47	19.47		19.89	
4d $\frac{3}{2}$	52.37	52.35	52.30			55 ^v 53.2 ^x 53.5 ^t 43.3 ^u 51.9 ^w
				52.1(3) ⁱ 53.5(3) ^a 57(3) ^r		
4d $\frac{5}{2}$	52.50	52.47	52.42		52.42	
5d $\frac{3}{2}$	108.57	108.33	107.68			110 ^x 110 ^v 107 ^w
				120(14) ^b		
5d $\frac{5}{2}$	108.87	108.63	107.98		107.7	
6d $\frac{3}{2}$	193.43	191.71	188.54			190 ^w 200 ^v
				206(14) ^b 176(10) ^a		
6d $\frac{5}{2}$	194.02	192.27	189.03		191.0	
7d $\frac{3}{2}$	312.43	305.77	297.43			317.3 ^x 306.1 ^w 320 ^v
				279(15) ^a 324(32) ^b		
7d $\frac{5}{2}$	313.41	306.40	297.44		308.1	
8d $\frac{3}{2}$	471.00	453.64	436.99			478.3 ^x 460.4 ^w 490 ^v
				449(50) ^a 502(39) ^b		
8d $\frac{5}{2}$	472.53	450.82	430.83		463.4	
9d $\frac{3}{2}$	674.91	638.79	610.11			681.8 ^x 658 ^w 710 ^v
				643(47) ^a 720(67) ^b		
9d $\frac{5}{2}$	677.12	640.64	611.74		664.6	
10d $\frac{3}{2}$	930.12	864.65	819.55			947 ^w 980 ^v 920 ^y
				971(35) ^b		
10d $\frac{5}{2}$	956.97	906.51	868.27		914.6	

TABLE III. (Continued).

State <i>nl j</i>	0 K	This work			Experimental	Other theoretical	
		410 K	600 K	Experimental		NCA ^s	Other
11 <i>d</i> $\frac{3}{2}$	1241.20	1133.30	1066.74				1300 ^y
11 <i>d</i> $\frac{5}{2}$	1271.58	1163.89	1098.10			1217.8	
12 <i>d</i> $\frac{3}{2}$	1611.14	1445.58	1352.05				1578 ^w
12 <i>d</i> $\frac{5}{2}$	1647.76	1474.48	1378.50		1650(150) ^b	1752.4	1590 ^y
13 <i>d</i> $\frac{3}{2}$	2050.45	1808.10	1680.81				2087 ^w
13 <i>d</i> $\frac{5}{2}$	2095.07	1832.64	1680.81		2120(400) ^b		2020 ^y
14 <i>d</i> $\frac{3}{2}$	2562.46	221.58	2053.24				
14 <i>d</i> $\frac{5}{2}$	2616.62	2213.96	2026.99				
15 <i>d</i> $\frac{3}{2}$	3152.96	2694.43	2479.54				
15 <i>d</i> $\frac{5}{2}$	3218.21	2735.28	2512.84				
16 <i>d</i> $\frac{3}{2}$	3827.38	3229.78	2961.43				
16 <i>d</i> $\frac{5}{2}$	3934.26	3403.68	3151.77				
17 <i>d</i> $\frac{3}{2}$	4591.28	3835.56	3507.30				
17 <i>d</i> $\frac{5}{2}$	4711.20	3930.21	3594.69	4460(220) ^d			4480 ^d
18 <i>d</i> $\frac{3}{2}$	5450.22	4526.04	4133.16				
18 <i>d</i> $\frac{5}{2}$	5588.26	4538.96	4116.10	5750(280) ^d			5310 ^d
19 <i>d</i> $\frac{3}{2}$	6409.87	5333.56*	4877.02*				
19 <i>d</i> $\frac{5}{2}$	6569.10	5469.04	5007.04	6900(340) ^d			6250 ^d
20 <i>d</i> $\frac{3}{2}$	7475.23	6406.13*	5921.31*				
20 <i>d</i> $\frac{5}{2}$	7690.98	6869.24*	6457.03*	7700(400) ^d			7290 ^d
4 <i>f</i> $\frac{5}{2}$	71.24	71.24	71.17				
4 <i>f</i> $\frac{7}{2}$	71.24	71.24	71.17			71.62	71 ^y
5 <i>f</i> $\frac{5}{2}$	137.17	136.91	135.83				
5 <i>f</i> $\frac{7}{2}$	137.17	136.45	134.17			137.4	140 ^y
6 <i>f</i> $\frac{5}{2}$	233.87	231.65	226.88				
6 <i>f</i> $\frac{7}{2}$	233.43	230.01	226.13			233.9	230 ^y
7 <i>f</i> $\frac{5}{2}$	367.25	358.69	346.89				
7 <i>f</i> $\frac{7}{2}$	359.89	354.53	346.11			367.3	370 ^y
8 <i>f</i> $\frac{5}{2}$	543.18	521.39	498.98				
8 <i>f</i> $\frac{7}{2}$	538.41	517.03	494.50			542.1	540 ^y
9 <i>f</i> $\frac{5}{2}$	767.67	723.31	686.18				
9 <i>f</i> $\frac{7}{2}$	765.37	721.46	683.77			767.0	760 ^y
10 <i>f</i> $\frac{5}{2}$	1046.09	967.37	910.88				
10 <i>f</i> $\frac{7}{2}$	1042.51	959.17	898.17			1044.0	1030 ^y
11 <i>f</i> $\frac{5}{2}$	1385.46	1257.86	1176.53				
11 <i>f</i> $\frac{7}{2}$	1380.45	1230.35	1134.77			1435.1	
12 <i>f</i> $\frac{5}{2}$	1790.90	1597.07	1484.69				
12 <i>f</i> $\frac{7}{2}$	1784.27	1469.18	1308.45			1788.1	

TABLE III. (Continued).

State <i>nl j</i>	This work			Experimental	Other theoretical	
	0 K	410 K	600 K		NCA ^s	Other
13f $\frac{5}{2}$	2268.56	1987.95	1837.45			
13f $\frac{7}{2}$	2260.06	1551.69	1305.62	2270(400) ^c		2260 ^y
14f $\frac{5}{2}$	2824.44	2433.93	2237.73			
14f $\frac{7}{2}$	2813.82	1854.86	1550.06	2640(450) ^c		2810 ^y
15f $\frac{5}{2}$	3464.60	2945.96	2699.08			
15f $\frac{7}{2}$	3480.15	2252.31	1881.47	3540(500) ^c		3450 ^y
16f $\frac{5}{2}$	4195.11	3528.60	3224.54			
16f $\frac{7}{2}$	4243.85	2687.96	2242.28			
17f $\frac{5}{2}$	5021.57	4193.36	3826.76			
17f $\frac{7}{2}$	5109.90	3183.39	2654.26			
18f $\frac{5}{2}$	5950.15	4961.98	4530.87			
18f $\frac{7}{2}$	6084.25	3782.02	3162.00			
19f $\frac{5}{2}$	6986.86	5887.95*	5401.17*			
19f $\frac{7}{2}$	7173.49	4762.75	4064.00			
20f $\frac{5}{2}$	8137.23	7181.52*	6703.47*			
20f $\frac{7}{2}$	8383.30	7948.16*	7668.90*			

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tively large to include in detail completely. We have, therefore, denoted by an asterisk states for which the BBR effect has not been completely accounted for. At this stage, the discrete states should be described as belonging to a "semicontinuum" spectrum³⁰ and the oscillator

strengths should be obtained by simple interpolation between the lower-*n* states values and a few "real-continuum" photoionization cross section values. Since in this case the states are close to the ionization threshold, contributions from the continuum should be included,

TABLE IV. Lifetimes of K (in nsec).

State <i>nl j</i>	0 K	This work		Experimental	Other theoretical	
		355 K	600 K		NCA ^h	Other
5s $\frac{1}{2}$	46.50	46.50	46.49		46.38	46.7 ⁱ 46 ^j
6s $\frac{1}{2}$	87.12	87.07	86.49	68(9) ^a	87.84	90.9 ⁱ 87 ^j 88 ^k
7s $\frac{1}{2}$	158.83	157.58	152.77	165(12) ^b	160.3	160 ^j
8s $\frac{1}{2}$	267.23	259.58	245.15	260(14) ^b	269.2	270 ^j
9s $\frac{1}{2}$	419.55	396.14	366.59	441(18) ^b	423.4	420 ^j
10s $\frac{1}{2}$	623.32	571.17	519.78	600(13) ^b	628.3	620 ^j
11s $\frac{1}{2}$	886.09	788.30	706.86	910(12) ^b	895.4	870 ^j
12s $\frac{1}{2}$	1215.42	1050.79	929.82		1227.0	1200 ^j
13s $\frac{1}{2}$	1618.96	1361.68	1190.43			1650 ^j
14s $\frac{1}{2}$	1633.68	1449.54	1312.19			2200 ^j
15s $\frac{1}{2}$	2644.04	2118.65	1816.07			
16s $\frac{1}{2}$	3358.56	2617.08	2217.71			
17s $\frac{1}{2}$	4105.30	3133.55	2635.09			
18s $\frac{1}{2}$	4916.01	3689.52	3084.20			
19s $\frac{1}{2}$	5712.63	4249.18*	3542.67*			
20s $\frac{1}{2}$	6846.00	5029.51*	4176.45*			
21s $\frac{1}{2}$	7961.21	6462.16*	5617.13*			
4p $\frac{1}{2}$	27.51	27.51	27.51	27.3(3.0) ^b 27.8(5) ^d		27.1 ⁱ 29 ^j
4p $\frac{3}{2}$	27.16	27.16	27.16	28(2) ^c 27.8(5) ^d 26.0(5) ^e	26.63	26.9 ⁱ 28 ^j
5p $\frac{1}{2}$	127.06	127.05	126.86			123 ⁱ 130 ^j
5p $\frac{3}{2}$	124.02	124.02	123.82	120(4) ^c 140.8(1.0) ^e 133(3) ^f	121.1	120 ⁱ 130 ^j
6p $\frac{1}{2}$	321.67	319.86	306.33			298 ⁱ 320 ^j
6p $\frac{3}{2}$	312.77	311.04	298.21	310(15) ^f	298.7	300 ⁱ 315 ^j
7p $\frac{1}{2}$	619.47	590.93	523.75			620 ^j
7p $\frac{3}{2}$	601.80	574.80	511.19		572.0	600 ^j
8p $\frac{1}{2}$	1040.23	920.67	772.58			1050 ^j
8p $\frac{3}{2}$	1010.51	897.50	756.60		957.4	1000 ^j
9p $\frac{1}{2}$	1607.54	1311.20	1061.84			1600 ^j
9p $\frac{3}{2}$	1561.31	1280.89	1042.70		1477.6	1550 ^j

TABLE IV. (Continued).

State <i>nl j</i>	0 K	This work		Experimental	Other theoretical	
		355 K	600 K		NCA ^h	Other
10p $\frac{1}{2}$	2345.84	1769.92	1396.11			2250 ^j
10p $\frac{3}{2}$	2279.41	1733.80	1374.56		2153.9	2200 ^j
11p $\frac{1}{2}$	3267.54	2295.11	1773.56			3150 ^j
11p $\frac{3}{2}$	3173.43	2250.66	1749.30		3002.4	3050 ^j
12p $\frac{1}{2}$	4402.92	2892.11	2196.43			4300 ^j
12p $\frac{3}{2}$	4279.78	2842.13	2170.58		4088.5	4200 ^j
13p $\frac{1}{2}$	5779.52	3601.98	2703.75			5650 ^j
13p $\frac{3}{2}$	5608.89	3539.03	2671.11			5500 ^j
14p $\frac{1}{2}$	7567.98	4524.24	3375.80			
14p $\frac{3}{2}$	7336.27	4445.57	3334.65			
15p $\frac{1}{2}$	9414.94	5239.54	3844.69			
15p $\frac{3}{2}$	9131.65	5156.16	3801.39			
16p $\frac{1}{2}$	11 597.40	6132.64	4453.73			
16p $\frac{3}{2}$	11 237.34	6036.33	4404.31			
17p $\frac{1}{2}$	14 145.22	7129.79	5130.52			
17p $\frac{3}{2}$	13 694.08	7018.94	5073.99			
18p $\frac{1}{2}$	16 630.73	8155.28	5847.12			
18p $\frac{3}{2}$	16 621.73	8153.09	5845.96			
19p $\frac{1}{2}$	19 628.10	9367.26*	6690.54*			
19p $\frac{3}{2}$	19 617.77	9364.89	6689.29			
20p $\frac{1}{2}$	23 069.96	10 863.09*	7752.66*			
20p $\frac{3}{2}$	23 058.04	10 860.43*	7751.28*			
3d $\frac{3}{2}$	45.24	45.24	45.24			40 ⁱ
				42(3) ^g		39 ^j
3d $\frac{5}{2}$	45.85	45.85	45.84		41.64	40.8 ⁱ
4d $\frac{3}{2}$	285.56	284.94	282.06			283 ⁱ
						280 ^j
4d $\frac{5}{2}$	291.18	290.54	287.54		284.4	286 ⁱ
5d $\frac{3}{2}$	767.41	748.75	691.82			
				610(90) ^b		710 ^j
5d $\frac{5}{2}$	769.63	750.83	693.55		720.1	
6d $\frac{3}{2}$	1180.58	1093.01	947.06			
				890(60) ^b		1050 ^j
6d $\frac{5}{2}$	1168.54	1082.65	939.47		1066.2	
7d $\frac{3}{2}$	1600.98	1399.34	1175.78			
				1210(100) ^b		1500 ^j
7d $\frac{5}{2}$	1577.12	1381.25	1163.41		1416.3	
8d $\frac{3}{2}$	2117.36	1755.38	1448.70			
				1590(130) ^b		1910 ^j
8d $\frac{5}{2}$	2080.88	1730.68	1432.54		1844.4	
9d $\frac{3}{2}$	2763.05	2182.80	1777.07			
				2040(300) ^b		2450 ^j

TABLE IV. (Continued).

State <i>nl j</i>	This work			Experimental	Other theoretical	
	0 K	355 K	600 K		NCA ^h	Other
$9d \frac{5}{2}$	2715.17	2153.68	1758.79		2332.0	
$10d \frac{3}{2}$	3555.50	2686.29	2161.45			3280 ^j
$10d \frac{5}{2}$	3492.12	2651.41	2140.35		3116.3	
$11d \frac{3}{2}$	4520.80	2373.54	2604.68			4260 ^j
$11d \frac{5}{2}$	4432.58	3229.01	2578.25		3821.2	
$12d \frac{3}{2}$	5668.12	3941.46	3103.01			5400 ^j
$12d \frac{5}{2}$	5574.14	3900.13	3080.90		5316.4	
$13d \frac{3}{2}$	6990.31	4690.54	3662.76			
$13d \frac{5}{2}$	6874.17	4647.19	3642.66			
$14d \frac{3}{2}$	8829.85	5667.30	4371.03			
$14d \frac{5}{2}$	8683.40	5635.04	4368.87			
$15d \frac{3}{2}$	10435.45	6540.14	5024.76			
$15d \frac{5}{2}$	10262.00	6578.93	5103.79			
$16d \frac{3}{2}$	13204.24	7881.61	5971.31			
$16d \frac{5}{2}$	12985.80	7270.61	5410.54			
$17d \frac{3}{2}$	15741.69	9156.89	6899.59			
$17d \frac{5}{2}$	15648.07	9025.95	6787.13			
$18d \frac{3}{2}$	19222.24	10872.70*	8135.15*			
$18d \frac{5}{2}$	19026.89	10495.55*	7795.59*			
$19d \frac{3}{2}$	24133.76	15192.10*	11723.72*			
$19d \frac{5}{2}$	23764.69	16354.88*	13002.69*			
$4f \frac{5}{2}$	70.65	70.62	70.48			
$4f \frac{7}{2}$	70.65	70.63	70.48		62.93	58 ^j
$5f \frac{5}{2}$	125.70	125.34	124.03			
$5f \frac{7}{2}$	125.69	125.37	124.02		113.5	105 ^j
$6f \frac{5}{2}$	205.22	203.41	198.65			
$6f \frac{7}{2}$	205.20	203.39	198.62		186.7	180 ^j
$7f \frac{5}{2}$	313.04	308.16	297.26			
$7f \frac{7}{2}$	314.00	308.10	296.99		287.4	275 ^j
$8f \frac{5}{2}$	456.74	442.75	422.67			
$8f \frac{7}{2}$	456.68	442.33	420.86		418.8	400 ^j
$9f \frac{5}{2}$	638.03	610.43	577.71			
$9f \frac{7}{2}$	637.93	607.35	568.77		587.8	560 ^j
$10f \frac{5}{2}$	863.58	815.30	765.80			
$10f \frac{7}{2}$	863.43	790.82	716.32		795.8	760 ^j
$11f \frac{5}{2}$	1137.66	1060.06	989.02			
$11f \frac{7}{2}$	1137.46	973.49	848.30		1047.3	1000 ^j
$12f \frac{5}{2}$	1465.08	1347.76	1249.64			
$12f \frac{7}{2}$	1467.78	1237.64	1075.16		1353.3	1280 ^j

TABLE IV. (Continued).

State <i>nl j</i>	This work			Experimental	Other theoretical	
	0 K	355 K	600 K		NCA ^a	Other
13 <i>f</i> $\frac{5}{2}$	1849.24	1683.59	1555.16			
13 <i>f</i> $\frac{7}{2}$	1852.98	1528.48	1319.53			
14 <i>f</i> $\frac{5}{2}$	2298.63	2074.98	1911.70			
14 <i>f</i> $\frac{7}{2}$	2305.02	1868.74	1607.74			
15 <i>f</i> $\frac{5}{2}$	2815.43	2525.78	2323.98			
15 <i>f</i> $\frac{7}{2}$	2826.23	2257.33	1936.86			
16 <i>f</i> $\frac{5}{2}$	3404.69	3050.52	2810.86			
16 <i>f</i> $\frac{7}{2}$	3420.68	2711.73	2328.20			
17 <i>f</i> $\frac{5}{2}$	4071.00	3670.70	3401.53			
17 <i>f</i> $\frac{7}{2}$	4093.75	3260.67	3816.87			
18 <i>f</i> $\frac{5}{2}$	4819.61	4473.91*	4225.39*			
18 <i>f</i> $\frac{7}{2}$	4850.25	4087.64*	3649.84*			

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probably in the same fashion as in the works of Farley and Wing,³¹ and Spencer and co-workers.³² We must remember, however, that as n increases, the probability of collisional depopulation of these states also increases, and the blackbody-radiation effect becomes only one of the competing processes.

B. Specific comparisons

Below we review the level of agreement between our predictions, the available experimental data, and other theoretical calculations. For the sake of convenience, we have grouped the relevant references at the end of each table. From the theoretical works we single out four, when available, as having direct relevance here. (i) The numerical Bates-Damgaard predictions of Lindgaard and Nielsen,¹¹ which are expected to be the closest to ours. (ii) The results of Anderson and Zilitis,¹⁰ who used experimental energies, numerical outward integration with a central-field potential, and power-expansion solutions at asymptotic distances. (iii) The CA-type calculations of Grudzev and Denisov³³ who also included polarization effects. (iv) The calculations of Gounand,³⁴ who used a different cutoff criterion for the double summation⁴ (cf. Sec. II).

The experimental energy term values were taken from Moore's tables.³⁵ They were complemented by more re-

cent tabulations, wherever available, and by extrapolations using the available quantum defects for other states.

1. Lithium

The experimentally available energy levels by Moore³⁵ and Johansson³⁶ do not clearly distinguish between the fine-structure states $j = \frac{1}{2}$ and $\frac{3}{2}$. Neither do the lifetime measurements, because the splittings are relatively small. We have thus used the multiplet centers of gravity. There exist a number of experimental works on the lifetimes of the lowest two s , p , and d states, most notably the one by Gaupp and co-workers,³⁷ claiming a high accuracy for $2p$. Our value is very close to it. Hansen has recently measured the lifetimes of a significant number of s and d states, at temperatures 620 and 720 K. Our predictions at the corresponding temperatures fall within his experimental errors for most of his values. The cutoff radius for the dipole matrix element [cf. Eq. (27)] was taken equal to $r_c = 1.4862$ a.u., the Hartree-Slater²⁷ prediction for the atomic core radius.

2. Sodium

Sodium is probably the most widely used alkali-metal atom by experimentalists, for studies of varying purposes. Though the recent tabulation of energy levels³⁸ distinguishes between the $j = \frac{1}{2}$ and $\frac{3}{2}$ states, the more systematic experimental lifetime investigations do not, and,

TABLE V. Lifetimes of Rb (in nsec).

State <i>nl j</i>	This work				Other theoretical	
	0 K	350 K	460 K	Experimental	NCA ^v	Other
6s $\frac{1}{2}$	45.21	45.21	45.21	46(5) ^a	51.47	51.3 ^w 51 ^x
7s $\frac{1}{2}$	86.59	86.55	86.39	88(6) ^a 91(11) ^b	97.72	99 ^w 96 ^x 98 ^y
8s $\frac{1}{2}$	158.34	157.09	155.24	154(7) ^a 153(8) ^c	178.4	170 ^x
9s $\frac{1}{2}$	266.36	258.77	252.52	258(13) ^c 245(50) ^d	299.5	290 ^x
10s $\frac{1}{2}$	417.84	394.68	381.40	430(20) ^e	470.5	460 ^x 427 ^z
11s $\frac{1}{2}$	620.16	568.81	545.24	532(32) ^a	698.3	680 ^x 628 ^z
12s $\frac{1}{2}$	879.83	784.09	746.35	764(46) ^a 770(150) ^d	993.6	1000 ^x 887 ^z
13s $\frac{1}{2}$	1207.57	1046.35	989.55			1400 ^x 1210 ^z
14s $\frac{1}{2}$	1609.28	1357.41	1276.46	1260(250) ^d		1850 ^x 1600 ^z
15s $\frac{1}{2}$	2092.32	1720.32	1609.28			2400 ^x 2070 ^z
16s $\frac{1}{2}$	2664.27	2138.50	1991.02	2190(500) ^d		2620 ^z
17s $\frac{1}{2}$	3332.67	2616.68	2426.14	2600(600) ^d		3260 ^z
18s $\frac{1}{2}$	4105.03	3163.33*	2923.31*	3300(700) ^d		4000 ^z
19s $\frac{1}{2}$	4988.76	3806.88*	3513.70*			4850 ^z
20s $\frac{1}{2}$	5991.36	5032.60*	4755.94*			5810 ^z
5p $\frac{1}{2}$	27.04	27.04	27.04	30(3) ^f 29.4(7) ^g 27.8(9) ^h 28.5(1.1) ⁱ		28.2 ^w 30 ^x
5p $\frac{3}{2}$	25.69	25.69	25.69	26.0(1.8) ^j 25.5(5) ^k 25.8(8) ^g 27.1(1.4) ^f 27.0(5) ^l 28.2(9) ^m	26.51	26.7 ^w 29 ^x
6p $\frac{1}{2}$	124.03	124.02	123.94	125(4) ^a 131(5) ⁱ		110 ^w 120 ^x
6p $\frac{3}{2}$	114.03	114.02	113.94	112(3) ^a 111(3) ⁿ 118(8) ^k 114(6) ^o 109(7) ⁱ	99.25	109 ^w 105 ^x

TABLE V. (Continued).

State <i>nl j</i>	0 K	This work		Experimental	Other theoretical	
		350 K	460 K		NCA ^v	Other
7 <i>p</i> $\frac{1}{2}$	277.39	275.51	272.30	272(15) ^a		255 ^w 260 ^x
7 <i>p</i> $\frac{3}{2}$	256.72	255.05	252.32	246(10) ^a 240(20) ^p 233(10) ^c	221.1	236 ^w 240 ^x
8 <i>p</i> $\frac{1}{2}$	496.36	479.79	464.42			480 ^x
8 <i>p</i> $\frac{3}{2}$	463.00	448.73	435.57	400(80) ^d	403.6	440 ^x
9 <i>p</i> $\frac{1}{2}$	798.84	739.22	702.35			770 ^x
9 <i>p</i> $\frac{3}{2}$	749.21	697.56	665.34	665(40) ^a	659.3	720 ^x
10 <i>p</i> $\frac{1}{2}$	1201.44	1058.64	991.75			1150 ^x
10 <i>p</i> $\frac{3}{2}$	1130.45	1005.60	946.20	1000(60) ^a	1001.9	1190 ^w 1100 ^x 1190 ^z
11 <i>p</i> $\frac{1}{2}$	1720.42	1443.69	1336.90			
11 <i>p</i> $\frac{3}{2}$	1622.84	1378.76	1282.73	1450(85) ^a	1443.6	1550 ^x 1710 ^z
12 <i>p</i> $\frac{1}{2}$	2369.41	1896.92	1739.45			2220 ^{aa}
12 <i>p</i> $\frac{3}{2}$	2238.14	1818.78	1675.83	1550(200) ^f 1970(110) ^a	1998.2	2250 ^x 2150 ^x 2400 ^z 2220 ^{aa}
13 <i>p</i> $\frac{1}{2}$	3167.98	2424.42	2204.15			3000 ^x
13 <i>p</i> $\frac{3}{2}$	2996.25	2332.14	2130.35			3230 ^z 2900 ^x
14 <i>p</i> $\frac{1}{2}$	4129.11	3027.35	2731.45			3900 ^x
14 <i>p</i> $\frac{3}{2}$	3904.52	2917.38	2644.72	2600(400) ^f		3950 ^{aa} 4230 ^z 3750 ^x
15 <i>p</i> $\frac{1}{2}$	5269.01	3707.57	3322.19			
15 <i>p</i> $\frac{3}{2}$	4989.02	3584.04	3226.81			5410 ^z
16 <i>p</i> $\frac{1}{2}$	6604.02	4471.09	3981.79			
16 <i>p</i> $\frac{3}{2}$	6246.36	4327.30	3872.65			6800 ^z
17 <i>p</i> $\frac{1}{2}$	8149.23	5321.22	4712.82			
17 <i>p</i> $\frac{3}{2}$	7708.48	5156.94	4589.45	6400(1300) ^f		7930 ^{aa} 8410 ^z
18 <i>p</i> $\frac{1}{2}$	9933.04	6765.67*	6043.17*			
18 <i>p</i> $\frac{3}{2}$	9398.54	6517.17	5843.19			10 200 ^z
19 <i>p</i> $\frac{1}{2}$	12 325.87	7772.86*	6862.02*			
19 <i>p</i> $\frac{3}{2}$	11 645.23	7555.89	6705.66			12 300 ^z
4 <i>d</i> $\frac{3}{2}$	82.27	82.27	82.27	83.4 ^a 86(6) ^s	85.01	78.7 ^w

TABLE V. (Continued).

State <i>nl j</i>	0 K	This work 350 K	460 K	Experimental	Other theoretical NCA ^v	Other
$4d \frac{5}{2}$	88.74	88.74	88.74	89.5 ^a 94(6) ^b		84 ^w 78 ^x
$5d \frac{3}{2}$	240.03	239.72	238.96	205(40) ^t	266.2	241 ^w
$5d \frac{5}{2}$	231.78	231.48	230.76	230(23) ^a		241 ^w 255 ^x
$6d \frac{3}{2}$	253.62	251.32	248.60			
$6d \frac{5}{2}$	243.72	241.58	239.10	237(15) ^a 285(16) ^c	295.1	300 ^x
$7d \frac{3}{2}$	331.08	323.65	318.01			
$7d \frac{5}{2}$	319.57	312.72	307.55	325(22) ^a 388(25) ^c	386.5	400 ^x
$8d \frac{3}{2}$	455.48	438.01	427.86			
$8d \frac{5}{2}$	440.90	424.77	415.39	421(25) ^a 515(30) ^c	532.4	560 ^x
$9d \frac{3}{2}$	625.98	591.42	574.65			800 ^z
$9d \frac{5}{2}$	606.96	574.94	559.32	568(35) ^a 565(120) ^u	722.8	750 ^x
$10d \frac{3}{2}$	847.17	785.96	759.87			
$10d \frac{5}{2}$	822.10	765.20	740.80	758(60) ^a 720(120) ^d	988.9	1040 ^x 1070 ^z
$11d \frac{3}{2}$	1126.33	1025.91	987.16			
$11d \frac{5}{2}$	1092.41	999.00	962.71	980(80) ^a 975(200) ^d	1311.3	1400 ^x 1410 ^z
$12d \frac{3}{2}$	1461.07	1307.36	1252.66			
$12d \frac{5}{2}$	1418.92	1275.25	1223.68	1250(300) ^d	1676.9	1820 ^x 1830 ^z
$13d \frac{3}{2}$	1866.14	1640.82	1565.86			
$13d \frac{5}{2}$	1813.48	1602.34	1531.45	1400(300) ^d		2300 ^x 2330 ^z
$14d \frac{3}{2}$	2358.95	2038.18	1937.41			
$14d \frac{5}{2}$	2275.11	1978.16	1883.86			2910 ^z
$15d \frac{3}{2}$	2911.81	2477.91	2347.81			
$15d \frac{5}{2}$	2809.39	2407.09	2285.10	3740(700) ^d		3610 ^z
$16d \frac{3}{2}$	3564.98	2992.39	2827.23			
$16d \frac{5}{2}$	3439.99	2908.58	2753.52			4400 ^z
$17d \frac{3}{2}$	5156.45	4572.38*	4387.62*			
$17d \frac{5}{2}$	4976.55	4436.00*	4263.25*			5300 ^z
$18d \frac{3}{2}$	5156.45	4572.38*	4387.62*			
$18d \frac{5}{2}$	4976.55	4436.00*	4263.25*	5300(1100) ^d		6320 ^z

TABLE V. (Continued).

State <i>nl j</i>	This work			Experimental	Other theoretical	
	0 K	350 K	460 K		NCA ^v	Other
4 <i>f</i> $\frac{5}{2}$	59.44	59.42	59.39			
4 <i>f</i> $\frac{7}{2}$	59.44	59.43	59.40		57.21	53 ^x
5 <i>f</i> $\frac{5}{2}$	106.12	105.86	105.48			
5 <i>f</i> $\frac{7}{2}$	106.09	105.83	105.44		101.1	96 ^x
6 <i>f</i> $\frac{5}{2}$	173.94	172.43	170.96			
6 <i>f</i> $\frac{7}{2}$	173.84	172.34	170.87		164.8	160 ^x
7 <i>f</i> $\frac{5}{2}$	267.11	262.31	258.76			
7 <i>f</i> $\frac{7}{2}$	266.91	262.12	258.57		252.8	245 ^x
8 <i>f</i> $\frac{5}{2}$	389.72	378.48	371.73			
8 <i>f</i> $\frac{7}{2}$	389.40	378.11	371.22		367.6	360 ^x
9 <i>f</i> $\frac{5}{2}$	546.08	524.10	512.81			
9 <i>f</i> $\frac{7}{2}$	545.58	522.94	510.89	550(80) ^u	515.6	500 ^x 506 ^z
10 <i>f</i> $\frac{5}{2}$	739.99	701.95	684.64			
10 <i>f</i> $\frac{7}{2}$	739.27	697.12	676.96	680(100) ^u	697.8	680 ^x 686 ^z
11 <i>f</i> $\frac{5}{2}$	976.06	915.84	891.18			
11 <i>f</i> $\frac{7}{2}$	975.08	888.73	852.70	900(140) ^u	918.2	890 ^x 904 ^z
12 <i>f</i> $\frac{5}{2}$	1258.24	1169.66	1136.35			
12 <i>f</i> $\frac{7}{2}$	1256.95	1088.05	1030.04		1190.0	1140 ^x 1170 ^z
13 <i>f</i> $\frac{5}{2}$	1590.78	1468.79	1425.91			
13 <i>f</i> $\frac{7}{2}$	1593.40	1377.01	1305.48	1620(240) ^u		1470 ^z
14 <i>f</i> $\frac{5}{2}$	1977.75	1822.43	1770.37			
14 <i>f</i> $\frac{7}{2}$	1979.43	1657.53	1557.50			1830 ^z
15 <i>f</i> $\frac{5}{2}$	2423.41	2270.20	2218.47			
15 <i>f</i> $\frac{7}{2}$	2423.88	1656.73	1478.75	1960(290) ^u		2240 ^z
16 <i>f</i> $\frac{5}{2}$	2937.08	2756.49	2696.82			
16 <i>f</i> $\frac{7}{2}$	2931.09	2639.78*	2550.70*			2710 ^z

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TABLE VI. Lifetimes of Cs (in nsec).

State <i>nl j</i>	This work			Experimental	Other theoretical	
	0 K	350 K	600 K		NCA ^{dd}	Other
7s $\frac{1}{2}$	48.17	48.17	48.16	49(4) ^a 47(5) ^a	57.35	56.8 ^{ee} 56 ^{ff} 53 ^{gg}
8s $\frac{1}{2}$	90.92	90.86	90.08	96(14) ^b 87(9) ^c	106.5	104 ^{ee} 100 ^{ff} 98 ^{hh}
9s $\frac{1}{2}$	164.30	162.82	157.28	231(35) ^b 147(15) ^c 167(3) ^d 160(8) ^e	191.8	192 ^{ee} 180 ^{ff} 167 ^d 189 ^{hh}
10s $\frac{1}{2}$	273.96	265.73	250.20	260(12) ^f 270(5) ^d	319.2	300 ^{ff} 271 ^d 324 ⁱⁱ
11s $\frac{1}{2}$	426.96	402.75	371.87	343(22) ^f 411(8) ^d	498.2	480 ^{ff} 416 ^d 509 ⁱⁱ
12s $\frac{1}{2}$	630.26	577.62	524.69	545(30) ^f 571(15) ^g	751.5	700 ^{ff} 610 ^d 758 ⁱⁱ
13s $\frac{1}{2}$	891.51	794.28	711.29	754(35) ^d		1000 ^{ff} 866 ^d 1080 ⁱⁱ
14s $\frac{1}{2}$	1217.67	1055.81	933.63	959(50) ^d		1450 ^{ff} 1267 ^d 1486 ⁱⁱ
15s $\frac{1}{2}$	1618.20	1367.09	1195.16			1900 ^{ff} 1984 ⁱⁱ
16s $\frac{1}{2}$	2093.07	1727.05	1495.31			2450 ^{ff}
17s $\frac{1}{2}$	2656.00	2146.34*	1843.53*			
18s $\frac{1}{2}$	3317.98	2645.63*	2263.28*			

TABLE VI. (Continued).

State <i>nl j</i>	This work			Experimental	Other theoretical						
	0 K	350 K	600 K		NCA ^{dd}	Other					
$6p \frac{1}{2}$	33.66	33.66	33.66	34 ^h		35 ^{ee}					
				34.0(6) ⁱ		38 ^{ff}					
						34 ⁱⁱ					
$6p \frac{3}{2}$	29.48	29.48	29.48	28(2) ^j	30.67	30.9 ^{ee}					
				30.5(7) ⁱ		34 ^{ff}					
				29.7(2) ^k		30 ⁱⁱ					
				29.8(2) ^l							
				32.7(3) ^m							
$7p \frac{1}{2}$	159.30	159.14	158.42	155(5) ⁿ		138 ^{ee}					
				165(6) ^o		160 ^{ff}					
				158(3) ^d		169 ^{gg}					
				158(5) ^p		155 ^{hh}					
				158(5) ^q		130 ⁱⁱ					
$7p \frac{3}{2}$	133.80	133.68	133.24	136(4) ^o	113	121 ^{ee}					
				136(4) ^p		106 ⁱⁱ					
				135(3) ^d		135 ^{ff}					
				122(2) ⁱ		110 ^{hh}					
				131(4) ^k		350(?) ^{gg}					
				118 ^r							
				125 ^s							
				111(6) ^t							
				135(1) ^m							
				134.0(2.8) ^u							
				134(3) ^y							
				$8p \frac{1}{2}$		363.32	360.74	350.89	330(30) ^v	264.9	331 ^{ee}
									307(14) ^u		390 ^{ff}
									334(21) ^w		293 ⁱⁱ
				$8p \frac{3}{2}$		318.47	316.74	309.47	310(5) ^k	264.9	267 ^{ee}
274(12) ^u	234 ⁱⁱ										
318(18) ^w	160 ^{jj}										
240(20) ^x	320 ^{ff}										
$9p \frac{1}{2}$	654.13	636.36	592.10	575(35) ^u	498.0	730 ^{ff}					
						526 ⁱⁱ					
$9p \frac{3}{2}$	593.97	580.18	543.29	502(22) ^u	498	615 ^{ff}					
				575(30) ^w		420 ⁱⁱ					
				390(30) ^l							
$10p \frac{1}{2}$	1050.77	985.02	877.23	920(50) ^y		1200 ^{ff}					
						837 ⁱⁱ					
$10p \frac{3}{2}$	976.60	921.00	826.47	900(40) ^y	825.6	1020 ^{ff}					
						670 ⁱⁱ					
$11p \frac{1}{2}$	1573.01	1407.73	1210.58			1800 ^{ff}					
						1236 ⁱⁱ					
$11p \frac{3}{2}$	1483.89	1337.77	1157.69		1264.9	1550 ^{ff}					
						992 ⁱⁱ					

TABLE VI. (Continued).

State <i>nl j</i>	0 K	This work		Experimental.	Other theoretical	
		350 K	600 K		NCA ^{dd}	Other
12p $\frac{1}{2}$	2241.49	1909.22	1598.98			2450 ^{ff} 1731 ⁱⁱ
12p $\frac{3}{2}$	2133.34	1832.43	1543.69		1817.2	2250 ^{ff} 1392 ⁱⁱ
13p $\frac{1}{2}$	3074.53	2495.41	2050.23			3350 ^{ff} 2340 ⁱⁱ
13p $\frac{3}{2}$	2945.59	2412.84	1993.45			3100 ^{ff} 1883 ⁱⁱ
14p $\frac{1}{2}$	4092.58	3177.04	2574.37			4450 ^{ff} 3071 ⁱⁱ
14p $\frac{3}{2}$	3937.27	3087.80	2516.12			4200 ^{ff} 2471 ⁱⁱ
15p $\frac{1}{2}$	5318.31	3975.10	3190.73			5700 ^{ff} 3919 ⁱⁱ
15p $\frac{3}{2}$	5128.16	3876.16	3129.51			5500 ^{ff} 3167 ⁱⁱ
16p $\frac{1}{2}$	6764.09	4923.41*	3934.28*			4930 ⁱⁱ
16p $\frac{3}{2}$	6530.19	4819.93	3878.47			3965 ⁱⁱ
17p $\frac{1}{2}$	8459.33	6149.40*	4937.43*			6078 ⁱⁱ
17p $\frac{3}{2}$	8174.00	6066.39*	4919.67*			4900 ⁱⁱ
18p $\frac{1}{2}$	10426.53	8047.46*	6640.36*			
18p $\frac{3}{2}$	10069.29	7869.89*	6538.96*			
5d $\frac{3}{2}$	909.15	909.15	908.74	890(90) ^a		952 ^{ee} 1164 ^{gg} 956 ⁱⁱ
5d $\frac{5}{2}$	1283.40	1283.39	1282.07	890(90) ^a	1110.5	1370 ^{ee} 1150 ^{ff} 1164 ^{gg} 1343 ⁱⁱ
6d $\frac{3}{2}$	58.16	58.14	58.02	57(15) ^z 60.0(2.5) ^e	69.89	64.5 ^{ee} 60.65 ⁱⁱ
6d $\frac{5}{2}$	58.39	58.38	58.26	60.7(2.5) ^e	69.89	65.5 ^{ee} 69 ^{ff} 60.5 ⁱⁱ
7d $\frac{3}{2}$	87.26	87.06	86.33	89(4) ^e 98(10) ^c		82.9 ⁱⁱ
7d $\frac{5}{2}$	88.06	87.87	87.16	88(9) ^e	102.9	100 ^{ff} 83.5 ⁱⁱ
8d $\frac{3}{2}$	138.45	137.38	135.02	152(3) ^d 154(5) ^e		124 ⁱⁱ
8d $\frac{5}{2}$	139.99	138.93	136.62		161.1	160 ^{ff} 125 ⁱⁱ

TABLE VI. (Continued).

State <i>nl j</i>	0 K	This work		Experimental	Other theoretical	
		350 K	600 K		NCA ^{dd}	Other
$9d \frac{3}{2}$	212.19	208.72	203.38	218(4) ^d		181 ⁱⁱ
$9d \frac{5}{2}$	214.70	211.28	205.85	95(10) ^{aa}	245.2	250 ^{ff} 183 ⁱⁱ
$10d \frac{3}{2}$	311.54	303.22	292.69	311(6) ^d		257 ⁱⁱ
$10d \frac{5}{2}$	315.29	307.04	296.52		357.3	350 ^{ff} 260 ⁱⁱ
$11d \frac{3}{2}$	440.22	423.52	405.13	428(12) ^d		352 ⁱⁱ
$11d \frac{5}{2}$	445.55	428.98	410.80		504.4	520 ^{ff} 357 ⁱⁱ
$12d \frac{3}{2}$	601.62	571.53	541.60	561(18) ^d 575(25) ^e		470 ⁱⁱ
$12d \frac{5}{2}$	608.87	579.34	550.38		687.4	477 ⁱⁱ
$13d \frac{3}{2}$	800.54	749.19	702.09	741(22) ^d 760(40) ^e		612 ⁱⁱ
$13d \frac{5}{2}$	809.84	761.46	717.98			920 ^{ff} 621 ⁱⁱ
$14d \frac{3}{2}$	1039.27	944.51	865.75	980(30) ^d		780 ⁱⁱ
$14d \frac{5}{2}$	1051.55	977.31	915.13			1170 ^{ff} 792 ⁱⁱ
$15d \frac{3}{2}$	1322.75	1168.56	1051.55	1175(38) ^{bb}		976 ⁱⁱ 1180 ^d 1573 ^{kk}
$15d \frac{5}{2}$	1335.77	1228.16	1143.04			990 ⁱⁱ
$16d \frac{3}{2}$	1662.32	1369.58	1162.65	1492(38) ^z		1207 ⁱⁱ 1443 ^d 1957 ^{kk}
$16d \frac{5}{2}$	1667.06	1520.45	1409.22			1224 ⁱⁱ
$4f \frac{5}{2}$	49.95	49.94	49.86			
$4f \frac{7}{2}$	50.09	50.09	50.00		44.93	49 ^{ff}
$5f \frac{5}{2}$	87.96	87.79	86.63			
$5f \frac{7}{2}$	88.10	87.92	86.97	92(8) ^w 95(6) ^w	76.78	85 ^{ff}
$6f \frac{5}{2}$	143.85	142.65	139.52			
$6f \frac{7}{2}$	143.95	142.76	139.64	150(10) ^w	123.8	140 ^{ff}
$7f \frac{5}{2}$	221.00	217.10	210.27			
$7f \frac{7}{2}$	221.04	217.17	210.39	230(20) ^w	189.2	220 ^{ff}
$8f \frac{5}{2}$	322.86	313.82	301.56			
$8f \frac{7}{2}$	322.81	313.84	301.67	330(30) ^w	274.8	320 ^{ff}
$9f \frac{5}{2}$	452.91	435.57	416.09			
$9f \frac{7}{2}$	452.72	435.54	416.20	470(30) ^w	385.1	450 ^{ff}
$10f \frac{5}{2}$	614.65	585.45	557.05			

TABLE VI. (Continued).

State <i>nl j</i>	This work			Experimental	Other theoretical	
	0 K	350 K	600 K		NCA ^{dd}	Other
10f $\frac{7}{2}$	614.27	585.35	557.18	635(30) ^w	521.1	610 ^{ff}
11f $\frac{5}{2}$	811.66	766.48	727.33			
11f $\frac{7}{2}$	810.94	766.22	727.41	830(5) ^{cc} 825(40) ^w	689.9	800 ^{ff}
12f $\frac{5}{2}$	1047.11	983.45	932.83			
12f $\frac{7}{2}$	1046.20	983.23	933.08	1060(25) ^{cc}	891.4	1030 ^{ff}
13f $\frac{5}{2}$	1324.21	1244.94 [*]	1185.29 [*]			
13f $\frac{7}{2}$	1323.32	1244.89	1185.83			
14f $\frac{5}{2}$	1647.22	1591.11 [*]	1546.99 [*]			
14f $\frac{7}{2}$	1645.94	1590.87 [*]	1547.54 [*]			
16f $\frac{5}{2}$	2652.47	2638.82 [*]	2623.17 [*]			
16f $\frac{7}{2}$	2645.27	2632.02 [*]	2616.80 [*]			
17f $\frac{5}{2}$	3196.33	3186.56 [*]	3173.75 [*]			
17f $\frac{7}{2}$	3186.98	3177.49 [*]	3165.00 [*]			

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therefore, their values should lie somewhere between our two numbers. For lifetimes we have, for lower states, the works by Gallagher and co-workers and by Kaiser, whereas Gounand and co-workers have been studying high- n states. Gaupp and co-workers³⁷ have also measured the lifetime of the $3p(j=\frac{1}{2})$ state with a high claimed accuracy; we refer to the latter work for an exhaustive comparison of their measured oscillator strength and various theoretical predictions. Our lifetime value is only slightly off theirs. In the other states, agreement between theory and experiment is fair. It should be noted, however, that there is no accord among different experimental values either. Of special interest here are the works of Spencer and co-workers, who studied the systematic BBR effects, as a function of temperature, on the lifetime³⁹ of the $19s$ and on the BBR-induced photoionization³² of the $15d$ states.

3. Potassium

The energy levels were taken from the recent tabulation of Corliss and Sugar.⁴⁰ Here again, the more systematic lifetime measurements do not distinguish between the fine-structure components of a state, and the lifetime value should lie somewhere between our two numbers. The only extensive experimental data were obtained by Gallagher and Cooke, and are in reasonable agreement with our values. It is interesting to notice here that the analytical CA results of Gruzdev and Denisov³³ are closer to ours than the numerical CA data of Lindgaard and Nielsen.¹¹

4. Rubidium

This has been a widely studied atom. The energy levels were taken from Moore's tables and from the more recent measurements of Johansson.⁴¹ The works of Marek and co-workers and of Gounand and co-workers are the more extensive ones. The former distinguish between the fine-structure components and thus make a systematic comparison with theory more meaningful. The predicted and measured lifetimes appear to be in an overall good agreement. The other theoretical values are in overall fair agreement with experiment.

5. Cesium

The energy levels were taken from Moore's tables and from an assortment of more recent works.⁴² Here again we have a wide range of experimental investigations and their overall agreement with our theoretical values is good, when they agree with one another. A very thorough comparison between all available techniques when applied to the Cs($5p$) state lifetime was given by Pace and Atkinson.⁴³ The lifetimes we obtained by using Norcross's potential¹⁶ were not significantly different. The predictions of the numerical CA approximation,¹¹ as well as the analytical CA values of Gruzdev and Denisov,³³ Happer,⁴⁴ and Gounand,³⁴ are all off for all s , p , and d states. The reason should be attributed to the significant size of the Cs atom core. This renders approximations that ignore the core obsolete.

Note added in proof. After submission of this paper W. S. Neil and J. B. Atkinson published lifetimes of S and D states in Cs in good agreement with our values [J. Phys. B 17, 693 (1984)].

ACKNOWLEDGMENTS

This work was supported at its early stage by the National Science Foundation, during the author's stay at The Johns Hopkins University, and at its final stage by a University of Toledo Faculty Research Award.

APPENDIX: NUMERICAL ASPECTS OF THE PROGRAM

Numerov's method of solution for Eq. (42) is expressed as

$$y_{j+1} - 2y_j + y_{j-1} = (h^2/12)(y_{j+1}'' + 10y_j'' + y_{j-1}'') + O(h^6) \quad (\text{A1})$$

which leads to [neglecting $O(h^6)$ terms]

$$a_{j+1}y_{j+1} - b_jy_j + a_{j-1}y_{j-1} = 0 \quad (\text{A2})$$

with

$$a_j = 1 - (h^2/12)p_j, \quad (\text{A3})$$

$$b_j = 2[1 + (5h^2/12)p_j]. \quad (\text{A4})$$

In the above we have used the notation $y_i = y(x_i)$ and $p_i = p(x_i)$.

It is known that the numerical solution obtained from (A2) is unstable when $p(x) > 0$. In the case of bound single-electron states $p(x)$ is always positive at very small distances (near the origin) and at very large distances. In addition, when a centrifugal "barrier" is present,⁴⁵ $p(x)$ is positive in the region of the barrier. The range $[x_0, x_N]$ is consequently subdivided into three regions:

- (a) $x \in [x_0, x_{j_1}]$, x_{j_1} is the first zero of $p(x)$ (lower region);
- (b) $x \in [x_{j_1}, x_{j_2}]$, x_{j_2} is the last zero of $p(x)$ (middle region); and
- (c) $x \in [x_{j_2}, x_N]$ (upper region).

In the lower and upper regions the Numerov method can become stable by applying the method of Gauss elimination without pivoting.⁴⁶ With this method, we use the boundary conditions

$$y_0 = y_N = 0 \quad (\text{A5})$$

to successively eliminate one unknown value of y_j from (A2). Specifically we have the following.

(a) *Lower region:*

$$y_j = u_j + v_j y_{j+1}, \quad (\text{A6a})$$

$$u_j = a_{j-1}u_{j-1} / (b_j - a_{j-1}v_{j-1}), \quad (\text{A6b})$$

$$v_j = a_{j+1} / (b_j - a_{j-1}u_{j-1}), \quad j = 1, 2, \dots, j_1 - 1 \quad (\text{A6c})$$

starting with

$$u_0 = v_0 = 0. \quad (\text{A7})$$

(b) *Upper region:*

$$y_j = u'_j + v'_j y_{j-1}, \quad j = j_2, j_2 + 1, \dots, N \quad (\text{A8a})$$

$$u_j = a_{j+1} u'_{j+1} / (b_j - a_{j+1} v'_{j+1}), \quad (\text{A8b})$$

$$v'_j = a_{j-1} / (b_j - a_{j+1} v_{j+1}), \quad j = N-1, N-2, \dots, j_2 \quad (\text{A8c})$$

starting with

$$u_N = v_N = 0. \quad (\text{A9})$$

(c) *Intermediate region:* the full relation (A2) is used in this case. The starting value $y_{j_1} = y(x_{j_1})$ at the first zero of $p(x)$ is taken equal to unity. The last value

$y_{j_2-1} = y(x_{j_2-1})$ is used as the starting value for the recursion relation of the upper region.

The adopted input parameters which were found optimal are

$$h = 0.1, \quad b = 3,$$

and

$$r_{\max} = (n^*)^2 \{ 1 + [1 + 5(10/n^*)^2]^{1/2} \}.$$

N was chosen to ensure a minimum of 10–20 calculation points per wave-function loop.

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